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High-Resolution Mapping and Modeling of Vehicle Emissions to Understand
Urban Air Quality Challenges

By

Katelyn Anna Yu

A dissertation submitted in partial satisfaction of the
requirements for the degree of

Doctor of Philosophy

in

Engineering - Civil and Environmental Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Robert A. Harley, Chair
Professor Inez Fung
Professor Cesunica Ivey

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Abstract

High-Resolution Mapping and Modeling of Vehicle Emissions to Understand Urban Air Quality Challenges

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University of California, Berkeley

Professor Robert A. Harley, Chair

The widespread utilization of vehicles in urban regions is a significant contributor to the pervasive air quality challenges faced by many cities globally. Among the pollutants emitted from vehicles, nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) play a pivotal role in influencing air quality. NO_x emission reduction has been a primary focus in air quality improvement efforts given the substantial contribution of NO_x to the formation of secondary pollutants with environmental and human health consequences, notably ozone and particulate matter ($\text{PM}_{2.5}$). While emissions have decreased significantly over the last few decades, difficulty constraining vehicle emissions has led to discrepancies among emission estimates derived from field campaigns, satellite observations, and models developed and used by government agencies. This research seeks to develop, assess, and implement a high-resolution emission inventory for motor vehicles. The broader objective of this work is to improve the understanding of spatial and temporal trends in vehicle emissions which is critical to developing effective air pollution control plans.

In alignment with these objectives, Chapter 2 assesses long-term (1990-2020) trends in vehicle emissions for the United States as a whole and more specifically for California. Unlike previous studies, which only considered emissions from light-duty gasoline and heavy-duty diesel vehicles, this research introduces a new category: light- and medium-duty diesel vehicles. Analysis of roadside spectrometer measurements reveals that this vehicle category exhibits the highest NO_x emission factors and the slowest rate of reduction over the past three decades. Assessment of on-road NO_x emission trends indicates a consistent pattern, with both the US and California experiencing $\sim 70\%$ decreases in NO_x emissions between 1990 and 2020. The relative reduction in diesel NO_x emissions has been larger in California (48%) than nationally (32%) since 2010. This is attributed to the rapid turnover of the diesel truck fleet in California, with older engines being replaced with more modern engines equipped with advanced emission control equipment. Emission estimates for California indicate convergence between gasoline and diesel source contributions to NO_x emissions, whereas the majority of on-road vehicle NO_x emissions in the US still come from diesel engines. Analysis of emission factor trends suggests diminishing returns in reducing gasoline emission factors, emphasizing the potential benefits of

improving emission control for light- and medium-duty diesel vehicles to maintain the downward trend.

Chapter 3 builds upon the emission trend analyses of Chapter 2 to assess the impact of emission control advancements on spatial patterns of NO_x emissions and concentrations in the Los Angeles metropolitan area. This chapter develops a high-resolution 1.3 km gridded motor vehicle emission inventory, characterized by higher overall NO_x emissions and a more pronounced drop-off on weekends attributable to the inclusion of the light- and medium-duty diesel vehicle category with higher emission factors. The new high-resolution inventory is used as input to the Weather Research and Forecasting model with Chemistry (WRF-Chem). Model results are evaluated using aircraft and TROPOMI satellite measurements of nitrogen dioxide (NO₂). This evaluation focuses on model skill in reproducing large observed decreases in atmospheric NO_x levels on weekends. The model showed comparable performance on weekdays and weekends, indicating appropriate day-of-week scaling and sector-based emissions distribution. This inventory suggests that on-road vehicles are responsible for 55-72% of NO_x emissions in the South Coast Air Basin, higher than the current amount (43%) attributed to vehicles in the management district planning inventory.

Chapter 4 explores the future impacts on secondary particulate matter (PM_{2.5}) resulting from vehicle-sourced NO_x emissions and analyzes how reductions in NO_x will influence the magnitudes and spatial distributions of PM_{2.5}, NO_x, and associated human health impacts. This chapter projects vehicle emissions from 2021 forward in time to 2030 and 2040 and expands the previously developed high-resolution NO_x emission inventory from Los Angeles to encompass the entire state of California. Estimated emissions are used to determine resulting patterns of decrease in NO_x and secondary PM_{2.5} using the Intervention Model for Air Pollution (InMAP). The resulting emissions are 62% higher in the fuel-based inventory in comparison to EMFAC in the baseline year 2021, with the largest discrepancy occurring in the light-/medium-duty diesel category. Predicted NO_x impacts remain concentrated near major freeways, while secondary PM_{2.5} impacts extend further from high-emission areas, likely due to transport and spatially variable atmospheric reaction rates. Significant decreases in secondary PM_{2.5} exposure are observed, particularly in the San Joaquin Valley and San Diego County. There is no clear pattern in the reduction of PM_{2.5} exposure by race, indicating that while reducing vehicle emissions may benefit all communities, additional measures are needed to address existing exposure disparities among different populations.

Key findings include the importance of addressing light-/medium-duty diesel engines as a significant NO_x emission source, given their persistently high emissions and slow rate of decrease. The fuel-based inventory performed well against observational data and estimated higher on-road emissions in comparison to regulatory inventories. This research provides a structure for how fuel-based emission inventories can be used to understand and target emission reduction efforts. Future studies should leverage new measurement technologies like the geosynchronous TEMPO satellite, model impacts on tropospheric ozone formation, evaluate impacts of reductions in primary PM_{2.5} emissions, and broaden the scope of the vehicle emission inventories developed in this work to understand source contributions to air pollution more broadly across the US and in other countries as well.

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Chapter 1: Introduction

1.1 Vehicle Contributions to Air Quality Challenges

Anthropogenic emissions from fossil fuel use pose a significant and multifaceted threat to the climate, the environment, and human health. The combustion of fossil fuels has increased dramatically amidst the acceleration of urbanization and heightened transportation demands globally with the growth of commerce, communication, and human activity.¹ Vehicles in particular have emerged as a major contributor to atmospheric pollution, with vehicular emissions assuming a central role in shaping the current state of air quality seen today in many major metropolitan areas. Urban centers characterized by dense populations and high concentrations of traffic often experience the highest concentrations of pollutants. Combustion of fossil fuels in internal combustion engines releases a complex mixture of pollutants, including carbon dioxide (CO₂), nitrogen oxides (NO_x = NO + NO₂), particulate matter (PM_{2.5}), carbon monoxide (CO), and volatile organic compounds (VOCs).²⁻⁵ There is an urgent need for comprehensive strategies to diminish the impact of these pollutants on air quality and climate change.

NO_x stands out as a particularly concerning component within the mix of vehicle emissions due to its role as a precursor to pollutants that pose a substantial threat to human and environmental health. In the atmosphere, NO_x reacts with VOCs in the presence of sunlight to create tropospheric ozone, which has negative impacts on plant growth and can exacerbate chronic respiratory conditions such as asthma.^{6,7} Acid rain formed through the reaction of oxidized NO_x with moisture can damage aquatic and soil ecosystems as well as result in significant economic costs.⁶ PM_{2.5} in the form of particulate nitrate is produced through the reaction of nitric acid and ammonia. Due to its danger to human health, PM_{2.5} has been at the forefront of air quality reduction initiatives. Its capacity to penetrate deep into the lungs underlies the observed associations between higher PM_{2.5} exposure and adverse impacts on respiratory, pulmonary, and cardiovascular health.⁸⁻¹³ Given the harmful consequences of pollutants resulting from NO_x chemistry, gaining a deeper insight into NO_x sources is becoming increasingly crucial for targeting emissions reductions effectively.

1.2 Emission Control Technology

The Clean Air Act, enacted in 1955 and subject to multiple revisions since, established ambient air quality standards and mandated compliance measures to meet these standards. As part of this framework, various regulations have been implemented and emission control technologies have targeted different major sectors over time to curb emissions. Initial regulatory focus centered on point-source power plants and these emissions have dropped dramatically due to the transition away from coal-sourced energy.^{14,15} As a result, the relative importance of the on-road sector has grown and there has been an increasing emphasis on curbing emissions from this sector. A crucial breakthrough in light-duty gasoline passenger vehicles occurred with the introduction of the three-way catalytic converter in the early 1980s, serving to reduce NO_x, CO,

and hydrocarbons in the exhaust. Pollutants are converted into less harmful gases using a catalyst; NO_x is reduced to N₂, CO is oxidized to CO₂, and unburned hydrocarbons (HC) are oxidized to CO₂ and H₂O.^{16,17} Emissions from light-duty gasoline passenger vehicles have decreased substantially as older vehicles have been phased out and replaced with newer and cleaner vehicles.^{18,19}

On the diesel side, however, the journey towards effective emission control systems has been marked by challenges alongside notable advancements. A key study by Yanowitz et al. highlights ineffective approaches to NO_x emission control on heavy-duty diesel vehicles.²⁰ Recent scandals exposing the evasion of emissions regulations by automotive manufacturers and transportation companies in diesel vehicles underscores the urgency of focused efforts on controlling emissions from diesel engines.^{21–23} In the last decade, the heavy-duty diesel truck sector has become a focal point for targeted regulatory interventions. A notable advance in this regard is the widespread adoption of Selective Catalytic Reduction (SCR) systems, which utilize a catalyst and a urea reagent introduced into the exhaust to facilitate reactions that convert NO_x into N₂ and water vapor. The implementation of SCR has been shown to reduce in-use NO_x emissions by >75% and represents a major advance in emissions control for heavy-duty diesel vehicles.^{24–29} SCR implementation offers a promising avenue for reducing harmful pollutant emissions and the transition is not limited to the US; major cities in North America, Europe, and Asia are incentivizing the adoption of SCR technology as part of broader efforts to mitigate adverse effects of vehicular emissions on urban air quality.^{30,31}

California's proactive stance on air quality and climate change has meant that state emission control rules and standards are often ahead of corresponding Federal measures. This offers a test case and insights into strategies that urban areas outside of California can consider implementing to address their air quality challenges. For example, the California Air Resources Board (CARB) oversees the Advanced Clean Cars Program, which sets aggressive tailpipe emissions standards for the light-duty vehicle fleet.³² This program also aims to increase zero-emission vehicle sales, aiming to reduce greenhouse gas emissions from light-duty vehicles by 50% by 2040. On the heavy-duty vehicle side, the Advanced Clean Truck program requires a higher percentage of zero-emission vehicle sales and has mandated fleet turnover to 100% 2010 and newer model year engines by 2023.³³ In particular, Los Angeles garners attention for air quality initiatives due to its susceptibility to smog formation, influenced by a unique confluence of geographic features, meteorological conditions, large vehicle-reliant population, and high-volume port activity.³⁴ California also has a long history of comprehensive air quality measurements, characterized by a dense network of routine air monitoring sites that is complemented by intensive research-oriented field campaigns.^{35,36} Improved remote sensing and emissions estimation methodologies for effective air quality management in California can lead to a better understanding of how dense urban regions should address NO_x emissions in the vehicle sector.

1.3 Emission Factor Measurements

The US on-road vehicle fleet is getting cleaner on average over time due to the adoption of emission control systems with improved durability and effectiveness. Emissions from the vehicle fleet exhibit an increasingly and now highly skewed distribution. This makes it harder to

quantify fleet-average emissions accurately because a small number of vehicles with either non-existent or malfunctioning emission control systems are contributing most of the emissions.²⁵ In addition, unusual vehicle operating conditions can lead to higher-than-expected emissions, which further complicates efforts to determine accurate and representative fleet-average emission factors.^{26,29,37,38} Current emission inventories developed by regulatory agencies incorporate distance-based emission factors (i.e., the mass of pollutant emitted per unit distance traveled) for individual vehicles derived from laboratory chassis dynamometer testing or on-board portable emission monitoring systems (PEMS).³⁹ While these methods can capture different driving modes and speed regimes, the number of vehicles that can be assessed is limited due to cost and labor constraints. This can lead to bias in fleet-average emission factors if high-emitting vehicles are missed or under-represented in the small populations of vehicles that are sampled.

As an alternative approach, roadside emission spectrometers operated across single-lane roads with lots of traffic can be used to measure emissions from many thousands of vehicles in just a week of fieldwork. The approach to calculating emission factors relies on a carbon balance: the mass of carbon emitted in the exhaust balances with the mass of carbon in the fuel entering the engine. Emission factors are calculated via Eq 1.1 using molar ratios of each pollutant to background-subtracted CO₂ derived for each passing vehicle:^{40,41}

$$F_{NO_x} = \frac{[NO] + [NO_2]}{[CO_2] + [CO] + [HC]} \cdot \frac{46}{12} \cdot w_c \quad \text{Eq 1.1}$$

F_{NO_x} is the emission factor (in units of grams of NO_x emitted per kg of fuel burned) and w_c is the weight fraction of carbon in the fuel (850-870 grams of carbon per kg of gasoline or diesel fuel). It is conventional to report NO_x emissions as if all emissions occur in the form of NO₂ (46 g/mol), even though direct NO_x emissions are mostly in the form of nitric oxide (NO). The contribution of HC to total carbon concentrations in the denominator of Eq 1.1 is usually negligible. By measuring on-road emission rates from a large sample of vehicles, these datasets can incorporate more engine types, vehicle conditions, and high emitters into calculated fleet-average emission factors. Using fuel-normalized emission factors also reduces variability due to differences in driving conditions (i.e., vehicle speed and acceleration) and vehicle weight in comparison to time- or distance-normalized emission factors.^{42,43}

1.4 Emission Inventories and Observations

Developing accurate vehicle emissions inventories is challenging due to the diverse nature of the vehicle fleet and the various driving and service conditions that vehicles operate under. This leads to significant spatial and temporal variation in vehicular NO_x emissions, making it difficult to constrain emission inventories. Past research utilizing top-down methods has exposed large uncertainties in existing inventory estimates and the understanding of the relative importance of various source sectors as contributors to overall emission totals. For instance, the 2011 DISCOVER-AQ aircraft field campaign measuring NO_x in the Baltimore-Washington region found that the EPA's Motor Vehicle Emission Simulator (MOVES) model overestimated mobile NO_x by 51-70% while the 2013 SEAC⁴RS aircraft campaign in the southeast US found this discrepancy to be up to a factor of 2.^{44,45} These studies show higher estimates of NO_x in the national MOVES emission model compared to field campaign values.

Satellite-based analyses of NO_x emissions have also demonstrated that MOVES-based estimates of emissions are decreasing more rapidly than observations suggest.⁴⁶ These studies underscore the limitations of emission models and highlight the need for developing and using alternative approaches to emissions quantification to estimate the largest sources of NO_x emissions accurately.

Advances in satellite-based remote sensing capabilities offer new opportunities for emissions quantification and verification. The newest satellites offer enhanced spatial and temporal resolution in the data they can provide. The Tropospheric Monitoring Instrument (TROPOMI) aboard the European Space Agency (ESA) Sentinel-5 Precursor satellite launched in 2017 has made significant strides towards higher spatial resolution with 3.5 x 5.5 km² reprocessed daily data available.^{47,48} Looking forward, the NASA Tropospheric Emissions: Monitoring of Pollution (TEMPO) satellite launched in 2023 orbits geosynchronously and will provide even higher resolution, with hourly measurements at 2 x 3 km² spatial resolution.⁴⁹

Fuel-based emission inventory methods provide a complementary perspective to emission model predictions and trends inferred from satellite data, providing critical insights into how emission control technologies have impacted NO_x concentrations in high-traffic urban areas. Both traditional chemical transport models (CTMs) like the Weather Research and Forecasting model⁵⁰ with chemistry (WRF-Chem) and reduced complexity models like the Intervention Model for Air Pollution⁵¹ (InMAP) are valuable tools for assessing the accuracy of emissions parameterizations. Transitioning to a higher-resolution emission inventory and air quality model is in line with recent improvements in satellite spatial resolution and the growing need to examine how changes to emissions will affect exposures to traffic-related air pollution in vulnerable near-roadway communities.

1.5 Research Objectives and Outline

The sprawling nature of US cities is characterized by extensive road networks and heterogeneous vehicle fleets, presenting a unique challenge in accurately assessing vehicle emissions. This dissertation provides a comprehensive analysis of emissions from the on-road sector with a particular focus on the U.S. and California, intending to piece apart and quantify the most important vehicle-fuel combinations as contributors to overall NO_x emissions. Through a focus on high-resolution emission mapping, this research aims to uncover underlying patterns and provide a foundation for targeted interventions and policies tailored to improving air quality in diverse urban environments. This in turn can contribute to the creation of healthier, more sustainable cities. To develop a better representation of the temporal and spatial patterns of NO_x and its impacts, the following chapters will cover the development, evaluation, and implementation of a high-resolution fuel-based inventory for motor vehicle emissions in California. The overarching questions of this work are as follows:

1. How has the implementation of emission control technologies driven trends in emission factors and total emissions between 1990-2020 at state and national scales?
2. What do high-resolution modeling of NO_x and satellite measurements of NO₂ for Los Angeles reveal about spatio-temporal distributions and the importance of various sources of NO_x?

3. How will future changes to the vehicle fleet affect spatial patterns of air pollution and human exposure to secondary PM_{2.5} that forms as a result of NO_x emissions from on-road vehicles?

The initial phase of the research is described in Chapter 2 and examines emission factor and NO_x emission trends from 1990 to 2020 at both national and California statewide levels. The research establishes and compares emissions trend lines for the US and California, utilizing taxable fuel sales data as a measure of vehicle activity and emission factors derived from roadside emission spectrometer and highway tunnel measurements. This study is unique in terms of separate modeling and highlighting of contributions to overall NO_x emissions from the comparatively unregulated light-/medium-duty diesel vehicle category. Despite a well-known emission cheating scandal, this vehicle category is typically overlooked or lumped together with other vehicle types in terms of its contribution to NO_x emissions.

Chapter 3 constructs a high-resolution emission inventory and integrates it into a chemical transport model (CTM) to see how model-derived NO₂ concentrations compare to satellite, aircraft, and ground observations from an air quality field study that took place in California in June of 2021. The inventory provides unusually high spatial resolution with 1.3 x 1.3 km² grid cells and incorporates emissions from light-/medium-duty diesel vehicles as a separately modeled category with high NO_x emission factors. Using the WRF-Chem model, the emission inventory and associated model predictions are evaluated in comparison to TROPOMI satellite data, vertical NO₂ concentration profiles measured by aircraft, and ground-based ambient air monitoring site data. The findings underscore the continuing dominant role of on-road vehicles as a source of NO_x and shed light on how emissions from specific vehicle sectors are at odds with predictions from California's EMFAC model. This comparison lays the groundwork for refining emission inventories in other urban areas, revealing key information about major sources of NO_x emissions in the greater Los Angeles metropolitan area.

In Chapter 4, the high-resolution emission mapping methodology is used to analyze the evolving magnitudes and spatial patterns of vehicle emissions and air pollution for future emission scenarios. This analysis focuses on changes in secondary PM_{2.5} and associated health effects in regions throughout California. This forward-looking analysis extends the inventory beyond the LA region to the full state, and projects magnitudes and spatial patterns of NO_x emissions for 2030 and 2040 to quantify projected changes in emissions. The emission projections are used with the InMAP model to reveal nuanced patterns of air quality improvements, emphasizing the importance of considering atmospheric transport and potential disparities in health benefits across the state. This chapter underscores the broader implications of the research by illuminating distinctions in the spatial distribution of NO_x emissions compared to secondary PM_{2.5} impacts, providing crucial information for devising targeted interventions to address urban air pollution.

Chapter 5 presents the conclusions, including key findings from Chapters 2-4 and the implications that these results have. The summary of results from the historical analysis, high-resolution inventory development, and future projections emphasizes the significance of the research in advancing the understanding of NO_x emissions and effects on air quality. The results also point out potential health benefits associated with continued reductions in NO_x emissions

and the need for targeted interventions. Chapter 5 concludes by detailing some natural directions that further research on this subject could take to build upon the results presented in this dissertation.

Chapter 2: Evaluation of Nitrogen Oxide Emission Inventories and Trends for On-Road Gasoline and Diesel Vehicles

This chapter is reproduced and adapted from:

Yu, K. A.; McDonald, B. C.; Harley, R. A. Evaluation of Nitrogen Oxide Emission Inventories and Trends for On-Road Gasoline and Diesel Vehicles. *Environ. Sci. Technol.* 2021, 55 (10), 6655–6664. <https://doi.org/10.1021/acs.est.1c00586>.

2.1 Introduction

Nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) are key atmospheric pollutants due to their effects on both human health and the environment. NO_x species are a major precursor in tropospheric ozone formation, which is a major component of smog and adversely affects human respiratory health.⁷ NO_x species are also precursors to acid rain and particle-phase aerosol nitrate.⁶ To manage and reduce these negative effects, it is important to identify the major sources of NO_x and to implement appropriate emission control efforts. NO_x is generated naturally from lightning and microbial processes in soil, but the largest source of emissions is anthropogenic and comes from the combustion of fossil fuels.⁵²

While 25% of the NO_x emissions in the United States were from electricity generation in 1999, programs to reduce point-source NO_x emissions and transition away from coal-generated electricity have caused stationary-source emissions to decrease significantly.^{14,15} As a result, the transportation sector is currently the largest contributor to NO_x emissions in the US.⁵² In the 1980s and 1990s, light-duty gasoline vehicles were the focus of mobile-source NO_x reduction efforts, which relied on the introduction and refinement of three-way catalytic converters that simultaneously reduce NO_x and oxidize carbon monoxide and hydrocarbons present in vehicle exhaust. NO_x emissions from light-duty gasoline vehicles have decreased steadily since the early 1990s, due to fleet turnover and increasingly effective and durable emission control systems installed on new gasoline engines.^{18,19,42}

Heavy-duty diesel vehicles became the dominant on-road source of NO_x in the early 2000s because of the decline in light-duty gasoline vehicle emissions and the rapid growth rate of diesel fuel sales.^{18,19} Despite the existence of increasingly stringent emission standards for new heavy-duty engines, efforts to control diesel NO_x emissions during the 1990s were ineffective.²⁰ More recently, diesel NO_x emission reduction goals have again been compromised by the re-installation of older heavy-duty engines in new “glider” trucks without inclusion of modern emission control systems and the use of “defeat devices” in light-duty diesel engines to disable NO_x emission control systems under real-world driving conditions.^{21–23} The implementation of stricter emission rules starting in 2004 led to declining diesel NO_x emissions from heavy-duty vehicles, and a further major advance started to take effect in 2010 with the introduction of selective catalytic reduction (SCR) systems to control NO_x from new diesel engines. Diesel SCR systems require the addition of a mixture of urea ($\text{NH}_2\text{-C(O)-NH}_2$) and water to the exhaust. The urea is converted to ammonia, which in turn undergoes a catalyzed

reaction with NO_x to form molecular nitrogen (N₂). SCR systems have been shown to reduce in-use NO_x emissions from heavy-duty diesel engines by >75%.²⁴⁻²⁹

National standards set limits on allowable emissions from new heavy-duty engines, but the in-use service lifetime for engines is long – over one million miles of driving may accrue over several decades of use.⁵³ To expedite reductions in emissions, California adopted rules to accelerate the turnover of the in-use diesel truck fleet. On-road heavy-duty diesel engines operating in California must meet stringent particulate matter (PM) emission standards by 2018, and NO_x emission standards by 2023.⁵⁴ These requirements have accelerated emission reductions in California compared to the rest of the country, but the implications for NO_x emissions trends due to emerging differences between national and California truck fleets have not been documented.

The extent to which the adoption of more effective NO_x control technology has affected emission trends in the US remains unclear. Field campaigns, satellite-based inventories, and chemical transport models have produced results showing discrepancies between the NO_x predicted by the MOVES emission model and observed emissions of NO_x. Field campaigns over the Baltimore Washington region and the southeast US found that MOVES overestimated NO_x in comparison to field campaign values.^{44,45} From 2011 to 2015, official US estimates of NO_x have also indicated more steeply decreasing emissions than the trends inferred from satellite observations of NO₂.⁴⁶ The contribution of free tropospheric NO₂ in satellite retrievals has been identified as a potential cause of the difficulty in capturing NO_x trends using top-down satellite observations.⁵⁵ Furthermore, NO_x lifetimes in US cities are changing, and this is a significant factor in satellite retrievals that remains generally unaccounted for.⁵⁶ To gain a more detailed understanding of NO_x emission trends over time, fuel-based methods can provide an important perspective that is complementary to emission model predictions and trends inferred from satellite data. Previous top-down studies have not separately evaluated light/medium-duty diesel emissions. Examining trends in on-road NO_x emissions in more recent years (i.e., since 2010) is of great importance, as the US and other countries in North America, Europe, and Asia have seen widespread adoption of SCR systems in new heavy-duty diesel vehicles.^{30,31} Furthermore, differences in diesel vehicle emission trajectories between California and the rest of the nation due to California's accelerated turnover of older heavy-duty trucks since 2010 have not been evaluated.

This study uses fuel sales data and on-road measurements of emissions factors from gasoline and diesel-fueled vehicles to estimate trends in NO_x emissions in both California and the United States as a whole. The goal of this analysis is to describe how NO_x emissions have changed over the period 1990-2020, with targeted consideration of how accelerated engine replacement requirements for heavy-duty trucks have caused California and national NO_x emission trends to diverge after 2010.

2.2 Methods

2.2.1 Fuel-Based Emission Inventory

A fuel-based emission inventory uses taxable fuel sales as a measure of on-road vehicle activity. NO_x is especially well-suited to a fuel-based analysis because the effects of vehicle-specific power on NO_x emission factors have become negligible.^{42,43} Fuel-based emission factors are expressed per unit mass of fuel burned, and a central aspect of the fuel-based inventory approach used in the present study is reliance on an extensive database of fuel-normalized emission measurements from real-world studies that feature large vehicle sample sizes. This differs from emission models that utilize vehicle activity data and often rely on measurements from relatively small samples of vehicles (i.e., laboratory testing and PEMS data), making it difficult to capture the increasingly important contribution to overall emissions from malfunctioning high-emitting vehicles.

Normalizing emission factors to fuel consumption reduces variability as a function of factors such as driving conditions and vehicle weight when compared to emission factors that are expressed per unit of time or unit distance traveled.^{42,43} That said, even fuel-normalized emission factors may be affected by driving cycles, for example, during extended periods of engine idling or when diesel exhaust temperatures are too low for SCR systems to be functional.^{29,57} However, these factors do not have major effects as extended idling accounts for <10% of diesel fuel consumption, the majority of diesel truck travel is on highways, and the effects of the driving cycle are small when emissions are fuel-normalized.^{19,20,58} For heavy-duty trucks in particular, engines manufactured from 1990-2004 may have higher than normal emissions when operating under very heavy engine loads, but this issue has been significantly reduced for newer engines.⁴²

For the state and national fuel sales used in this study, on-road gasoline sales were obtained from taxable fuel sales reports, and on-road diesel sales were obtained from distillate fuel sales surveys.⁵⁹⁻⁶¹ Separate California and 49-state NO_x emission inventory estimates were constructed, and results were then combined to arrive at national emission totals. We assumed uncertainties of ±3 and ±5% for national gasoline and diesel fuel sales, respectively, and ±5 and ±10% for sales of gasoline and diesel fuel in California, respectively.¹⁹ Uncertainties in fuel use and NO_x emission factors were propagated through emission inventory calculations.

2.2.2 NO_x Emission Factors

NO_x emission factors for light-duty gasoline, light/medium-duty diesel, and heavy-duty diesel vehicles for each year were derived from on-road studies referenced by McDonald et al.,¹⁹ with new results added from field measurements conducted since 2010. Measurement techniques used in light/medium-duty vehicle emission studies include remote roadside emission spectrometers that measure ratios of various pollutants to CO₂ within the exhaust plumes of individual vehicles as they drive by.^{42,62-64} Heavy-duty diesel emission factor measurements were made using plume capture methods that integrate time-varying concentrations of gaseous and particle-phase pollutants present in the exhaust plumes of individual trucks.^{25,28} For all the emission measurement techniques used here, the general underlying analytical principle used is

that measurements of NO_x concentrations and their relative ratios to CO₂ are combined using a carbon balance to derive emission factors that are expressed per unit mass of fuel burned.

For the light/medium-duty vehicle fleet, there have been a series of new datasets from recent on-road studies conducted in Van Nuys,⁶⁵ West Los Angeles,^{62–64,66} and Lynwood⁶⁷ in California, as well as elsewhere in the US in Tulsa,^{64,68–70} Denver,^{64,71} and Chicago^{42,72} (Table 2.1). Light/medium-duty vehicle sample sizes are large, with typically 15,000 to 30,000 vehicles observed during a sampling period. On the heavy-duty diesel side, newer results come from recent on-road emissions studies conducted in Anaheim,⁷³ Redding,²⁸ and Oakland²⁵ in California (Table 2.2). The sample size of vehicles measured for NO_x at the Oakland site under highway driving conditions was 557 vehicles for the 2010 fleet and between 1100 and 1200 vehicles for the 2014, 2015, and 2018 fleets.²⁵ There have been few large-scale on-road studies of diesel truck emissions outside of California since 2005. Separate emission factor trends were derived from the on-road emission data for California and the rest of the US, as there are important differences in national and California emission control programs.

Table 2.1: Measurements of NO_x emission factors from on-road studies of light/medium-duty vehicles

Year	Location	Gasoline EF (g/kg)	Diesel EF (g/kg)	Study Period	Reference
1992	Baltimore, MD	10.4	-	6/18/92 - 6/24/92	Bishop et al. 1996
1994	Oakland, CA	10.7	-	Summer	Kean et al. 2002
1995	Oakland, CA	9.6	-	Summer	Kean et al. 2002
1995	Van Nuys, CA	11.0	-	n/a	Bishop et al. 2012
1995	Denver, CO	19.0	-	12/11/95 - 1/13/96	Stedman et al. 1997
1996	Oakland, CA	9.0	-	Summer	Kean et al. 2002
1997	Oakland, CA	9.0	-	7/21/97 -7/24/97, 7/31/97 - 8/5/97	Ban-Weiss et al. 2008
1997	Chicago, IL	8.5	-	9/15/97 - 9/19/97	Bishop and Stedman, 2008
1997	Denver, CO	13.2	-	12/20/96 - 1/31/97	Stedman et al. 1998
1998	Chicago, IL	8.7	-	9/21/98 - 9/24/98	Bishop and Stedman, 2008
1999	Oakland, CA	6.6	-	Summer	Kean et al. 2002
1999	West LA, CA	9.9	21.7	11/9/99 - 11/13/99	Bishop and Stedman, 2008
1999	Riverside, CA	7.4	20.0	6/28/99 - 7/7/99	Pokharel et al. 2003
1999	San Jose, CA	6.4	21.5	10/27/99 - 10/29/99	Bishop et al. 2010
1999	Chicago, IL	8.1	-	9/20/99 - 9/23/99	Bishop and Stedman, 2008
1999	Phoenix, AZ	11.8	28.6	11/15/99 - 11/19/99	Bishop and Stedman, 2008
1999	Denver, CO	12.9	-	1/14/99 - 2/1/99	Bishop and Stedman, 2008
2000	Riverside, CA	8.7	22.3	5/30/00-6/6/00	Pokharel et al. 2003
2000	Chicago, IL	6.8	-	9/11/00 - 9/14/00	Bishop and Stedman, 2008
2000	Phoenix, AZ	8.1	26.0	11/13/00 - 11/17/00	Bishop and Stedman, 2008
2000	Denver, CO	10.7	30.1	12/30/99 - 1/14/00	Bishop and Stedman, 2008
2001	Oakland, CA	5.6	-	Summer	Kean et al. 2002
2001	West LA, CA	8.4	23.0	10/15/01 - 10/19/01	Bishop and Stedman, 2008
2001	Riverside, CA	8.1	23.1	6/6/01 - 6/13/01	Pokharel et al. 2003
2001	Denver, CO	10.1	33.8	1/5/01 - 1/8/01	Bishop and Stedman, 2008
2002	Chicago, IL	5.6	-	9/16/02 - 9/20/02	Bishop and Stedman, 2008
2002	Phoenix, AZ	6.3	24.0	11/18/02 - 11/22/02	Bishop and Stedman, 2008
2003	West LA, CA	6.5	22.0	10/27/03 - 10/31/03	Bishop and Stedman, 2008
2003	Denver, CO	9.2	34.1	12/31/02 - 1/7/03	Bishop and Stedman, 2008
2003	Tulsa, OK	5.7	-	9/8/03 - 9/12/03	Bishop and Stedman, 2008

2004	Chicago, IL	5.1	-	9/20/04 - 9/23/04	Bishop and Stedman, 2008
2004	Phoenix, AZ	4.4	23.2	11/15/04 - 11/19/04	Bishop and Stedman, 2008
2005	West LA, CA	4.8	23.5	10/17/05 - 10/21/05	Bishop and Stedman, 2008
2005	Denver, CO	7.3	31.9	1/8/05 - 1/11/05	Bishop and Stedman, 2008
2005	Tulsa, OK	4.0	-	9/19/05 - 9/23/05	Bishop and Stedman, 2015
2006	Oakland, CA	3.0	-	7/18/06 - 8/10/06	Ban-Weiss et al. 2008
2006	Chicago, IL	2.7	-	9/12/06 - 9/15/06	Bishop and Stedman, 2008
2006	Phoenix, AZ	3.5	21.2	11/13/06 - 11/17/06	Bishop and Stedman, 2008
2007	Denver, CO	5.4	25.4	1/10/07 - 2/27/07	Bishop and Stedman, 2008
2008	West LA, CA	5.8	-	3/17/08 - 3/21/08	Bishop et al. 2010
2008	San Jose, CA	4.1	-	3/4/08 - 3/7/08	Bishop et al. 2010
2008	Fresno, CA	4.4	-	3/8/08 - 3/14/08	Bishop et al. 2010
2010	Oakland, CA	1.9	-	7/6/2010-7/15/2010	Dallmann et al. 2013
2010	Van Nuys, CA	3.9	22.8	8/12/2010-8/16/2010	Bishop et al. 2012a
2010	Van Nuys, CA	3.3	-	8/12/10-8/16/10	Bishop et al. 2012a
2013	West LA, CA	3.3	20.8	4/27/13-5/4/13	Bishop and Stedman, 2015
2013	Denver, CO	3.8	26.6	12/12/13 - 12/13/13, 1/4/14	Bishop and Stedman, 2015
2013	Tulsa, OK	2.2	19.0	9/30/13 - 10/4/13	Bishop and Stedman, 2015
2014	Chicago, IL	2.5	-	9/8/14-9/13/14	Bishop et al. 2016
2014	Chicago, IL	2.2	-	9/8/14 - 9/13/14	Bishop and Haugin, 2018
2015	West LA, CA	3.3	16.4	3/28/15 - 4/3/15	Bishop and Stedman, 2016
2015	Denver, CO	2.6	24.2	1/13/2016	Unpublished
2015	Tulsa, OK	2.3	-	9/14/15 - 9/18/15	CRC Report No. E-106
2016	Chicago, IL	1.9	18.4	9/12/16 - 9/21/16	Bishop and Haugin, 2018
2017	Tulsa, OK	2.4	-	9/11/2017 - 9/15/2017	CRC Report No. E-123
2018	Lynwood, CA	3.4	20.6	5/7/2018 - 5/9/2018	CRC Report No. E-124
2018	Lynwood, CA	2.7	22.0	5/10/2018 - 5/12/2018	CRC Report No. E-124
2018	Denver, CO	2.5	22.0	12/15/17 - 1/18/18	CRC Report No. E-123
2018	Chicago, IL	1.4	14.7	9/10/18 - 9/15/18	CRC Report No. E-123
2019	Tulsa, OK	1.5	13.3	9/9/19 - 9/13/19	CRC Report No. E-123

*NO_x EF in NO₂-equivalence

Table 2.2: Measurements of emission factors from on-road studies of heavy-duty vehicles

Year	Location	NO_x EF (g/kg)	Error Bar	Study Period	Reference
1992	Fort McHenry	32.1	3.7	6/18/92-6/24/92	Bishop et al. 1996
1992	Tuscarora, PA	35.2	3.1	9/2/92-9/8/92	Bishop et al. 1996
1997	Oakland, CA	57.0	7.0	7/21/97 - 7/24/97, 7/31/97 - 8/5/97	Ban-Weiss et al. 2008
1997	Anaheim, CA	32.0	1.3	8/25/97 - 8/29/97	Bishop et al. 2012b
1997	Raleigh, NC	45.0	4.0	6/16/97 -6/19/97	Jimenez et al. 2000
1997	Chassis	34.6	0.9	n/a	Yanowitz et al. 2000
1999	Golden, CO	40.0	3.0	5/13-5/14/99	Burgard et al. 2006
2002	Pittsburgh, PA	43.0	11	November 2002 (2 weeks)	Grieshop et al. 2006
2005	Golden, CO	34.5	2.2	8/17/05-8/24/05	Burgard et al. 2006
2005	Nogales, AZ	36.0	3.0	March 2005 (5 days)	Wood et al. 2005
2006	Oakland, CA	40.0	3.0	7/18/06 - 8/10/06	Ban-Weiss et al. 2008
2008	Anaheim, CA	27.3	0.6	4/4/08, 4/7/08 - 4/10/08	Bishop et al. 2012b
2009	Anaheim, CA	25.4	1.0	4/27/09 - 5/1/09	Bishop et al. 2012b
2010	Oakland, CA	31.3	1.6	n/a	Preble et al. 2019
2010	Oakland, CA	28.0	1.5	July 2010	Dallmann et al. 2012
2010	Anaheim, CA	24.5	1.4	4/26/10 - 4/30/10	Bishop et al. 2012b
2012	Anaheim, CA	19.9	0.3	9/24/12 - 9/28/12	Bishop et al. 2013
2013	Redding, CA	20.3	1.4	5/6/13 - 5/10/13	Bishop et al. 2015
2014	Oakland, CA	16.3	0.9	n/a	Preble et al. 2019
2015	Oakland, CA	15.0	0.9	n/a	Preble et al. 2019
2015	Redding, CA	22.1	1.4	4/8/15-4/10/15	Haugen and Bishop, 2017
2015	PEMS Composite	20.0	n/a	n/a	Jiang et al. 2018
2017	Anaheim, CA	12.4	0.6	3/20/17-3/23/17	Haugen et al. 2018
2017	Redding, CA	18.6	1.2	4/10/17-4/14/17	Haugen and Bishop, 2018
2018	Oakland, CA	13.2	1.0	n/a	Preble et al. 2019

*NO_x EF in NO₂-equivalence

National and California light-duty gasoline vehicle NO_x emission factor trends were derived using quadratic fits to the measured fleet averages in each category. When applied over such a long period, linear fits to the data are not able to represent the decreasing absolute rate of reduction in emission factors in recent years. On the other hand, a linear regression was employed for the light/medium-duty diesel trend due to the significantly slower rate of decline in emission factors. Uncertainties in light-duty emission factors were calculated using 95% confidence intervals. For heavy-duty vehicles, available on-road data for NO_x emission factors are dominated by results of studies conducted in California. Using the data from studies conducted in 2004 or later, we derived a linear fit to the emission factor trendline for the heavy-duty diesel truck fleet operating in California. While a previous fuel-based inventory¹⁹ assumed a trend of decreasing heavy-duty NO_x emission factors starting in 1997, this study assumes a constant NO_x emission factor for all years before 2004. This is the year after which all new trucks were required to meet more stringent NO_x regulations.⁷⁴ Heavy-duty NO_x emission factors were assumed to remain constant at pre-control levels prior to 2004, as there was little, if any, in-use emission benefit from new-engine NO_x standards that were in effect in earlier years, as shown by chassis dynamometer studies on pre-2000 heavy-duty diesel engines.²⁰

Heavy-duty diesel NO_x emission factors are assumed to be the same nationally and in California until 2010, when California began to require accelerated turnover of the legacy truck fleet in favor of newer SCR-equipped trucks. Due to a lack of recent NO_x emission factor data for heavy-duty diesel trucks operating in states other than California, NO_x emission factors for the national truck fleet were generated by reweighting available California NO_x emission factors using age distributions from the California EMFAC and national MOVES models. In 2018, the percentage of the total VMT traveled by model year 2010 or newer trucks was 57% in the US and 74% in California (Figure 2.1). The reweighting was done by determining how much the fleet-average NO_x emission factor would differ between the US and California truck fleets in calendar year 2018 using contemporary measurements of NO_x emission factors as a function of engine model year from Preble et al.²⁵ The model-year-resolved emission factor data was then weighted by age-resolved truck travel distributions specified in the national MOVES and California EMFAC models. The resulting ratio of fleet-average NO_x emission factors for the national and California truck fleets in 2018 was 1.35. The national emission factor trendline was constructed by moving the California emission factor trendline up by 35% in 2018 and projecting back in time to 2010 when the difference between California and national truck fleets was negligible.

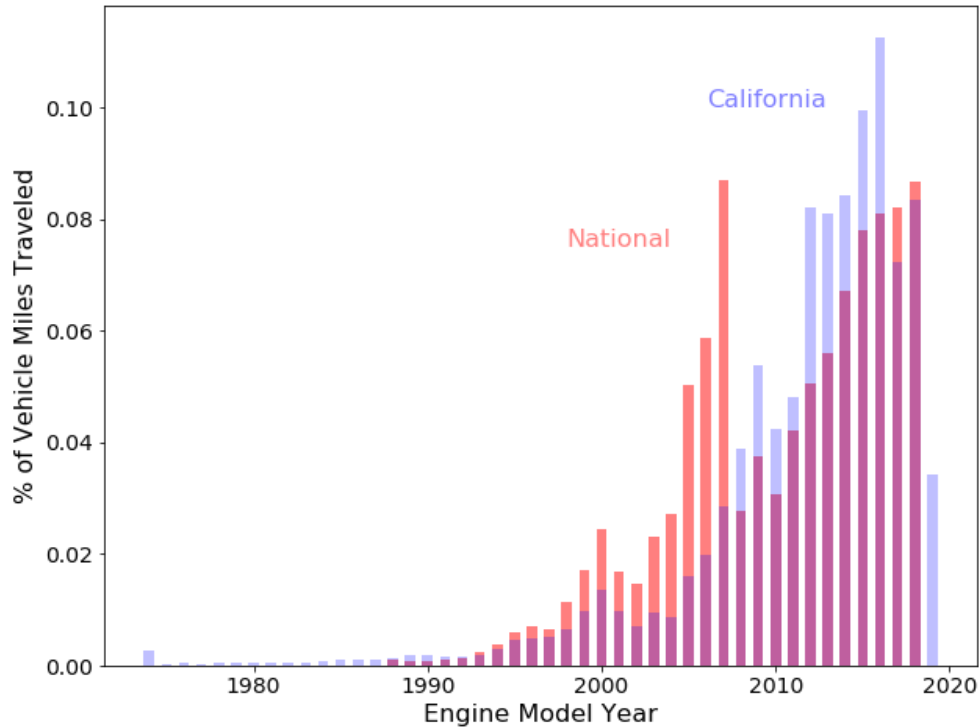


Figure 2.1: EMFAC (California) and MOVES (national) engine model year distribution comparison by vehicle miles traveled for the 2018 fleet

The uncertainties in light-duty gasoline and diesel vehicle NO_x emission factors are 95% confidence intervals of the regression trendlines. The uncertainty in heavy-duty vehicle NO_x emission factors for California is a 95% confidence interval for the post-2004 regression analysis, with the error for pre-2004 years set at the 2004 value. Heavy-duty diesel emission factors for the national truck fleet are assigned the same relative uncertainty as the emission factors for California, implying larger absolute uncertainties after 2010 since emission factors are higher for the national truck fleet.

2.2.3 NO_x Emissions Due to Cold Engine Starting

Cold start increments were estimated for California using cold start emission percentages derived from the EMFAC model for each calendar year since 2000, the earliest calendar year for which EMFAC predictions are available. Cold start fractions of total gasoline emissions ranged from 18 to 26% in EMFAC, while national cold start percentages derived from the MOVES model indicated a much larger increase from 10% in 1990 to 37% in 2018, with comparable cold start increments predicted for both NO_x and CO emissions. We expect cold start increments for CO to be higher than for NO_x ⁷⁵ and therefore chose to use the lower EMFAC-derived estimates of cold start fractions in estimating national start-related emission increments as well. There are no explicit uncertainty estimates for cold start-related emissions included in the emission models. Assuming the relative estimate of cold start impacts in the EMFAC model is reasonable, engine starting is a minor source of uncertainty in the overall on-road vehicle NO_x emission inventory. Further investigation of reasons for the disagreement between EMFAC and MOVES model estimates of cold start impacts is recommended but is beyond the scope of the present study.

2.2.4 Trends in Measured Ambient NO_x Concentrations

US and California trends were calculated using the light-duty gasoline emission factors for gasoline fuel sales and light/medium-duty and heavy-duty diesel emission factors for diesel fuel sales. Light/medium-duty diesel vehicles are estimated to account for 25-30% of total on-road diesel fuel consumption.⁷⁶ We used the estimated diesel fuel consumption by vehicle type from EMFAC to apportion fuel sales to light/medium-duty and heavy-duty diesel engines and calculate emissions separately for these vehicle weight categories. Cold start percentages from EMFAC were used to estimate appropriate increments to gasoline-source NO_x emissions, and EMFAC and MOVES estimate for emissions from transit and school buses were added to the diesel-source NO_x emission inventory.

Trends in measured concentrations of NO_x in the ambient air of two major urban areas (Los Angeles and Houston) were used as points of comparison for the NO_x emission trends developed in the present study. Ambient concentration measurements were obtained from the US EPA Air Quality System database.³⁶ The sensors selected were those that had at least 20 years of data during the 1990-2020 period (Figure 2.2, Figure 2.3). To focus the comparison between on-road emissions and ambient measurements, the raw ambient data were filtered to include only measurements made from 5:00-9:00 AM local standard time on weekdays. The daily peaks in NO_x concentrations observed during this timeframe are dominated by contributions from on-road vehicular sources, at a time when photochemical activity and vertical mixing rates are generally low.^{77,78} Fuel sales estimates for Los Angeles and gasoline sales estimates in Houston were taken from McDonald et al.,⁷⁹ updated for calendar years after 2010 using the relative increase in statewide fuel sales since 2010. To estimate diesel fuel sales in Houston, a 2002 inventory of carbon emissions from McDonald et al.⁸⁰ was converted to a volume of fuel and similarly scaled to other years using relative changes in statewide diesel fuel sales data for Texas. Uncertainties in fuel sales for the Los Angeles area were ±9-13% and ±11-15% for gasoline and diesel fuel, respectively.⁷⁹ For Houston, uncertainties of ±15-17% for gasoline fuel sales and ±14% for diesel fuel sales were used.^{79,80} Uncertainties in ambient data were not factored into the calculations due to the large number of measurements used to compute annual weekday morning averages of the relevant NO_x concentrations.

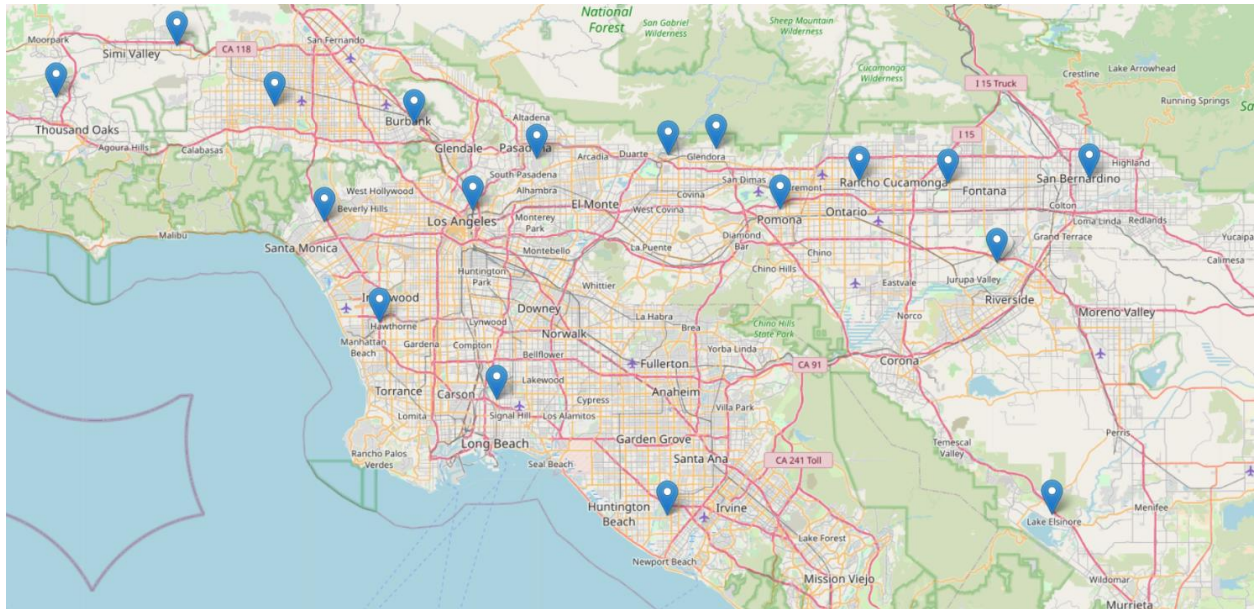


Figure 2.2: Long-running ambient air quality sensor locations in Los Angeles, CA

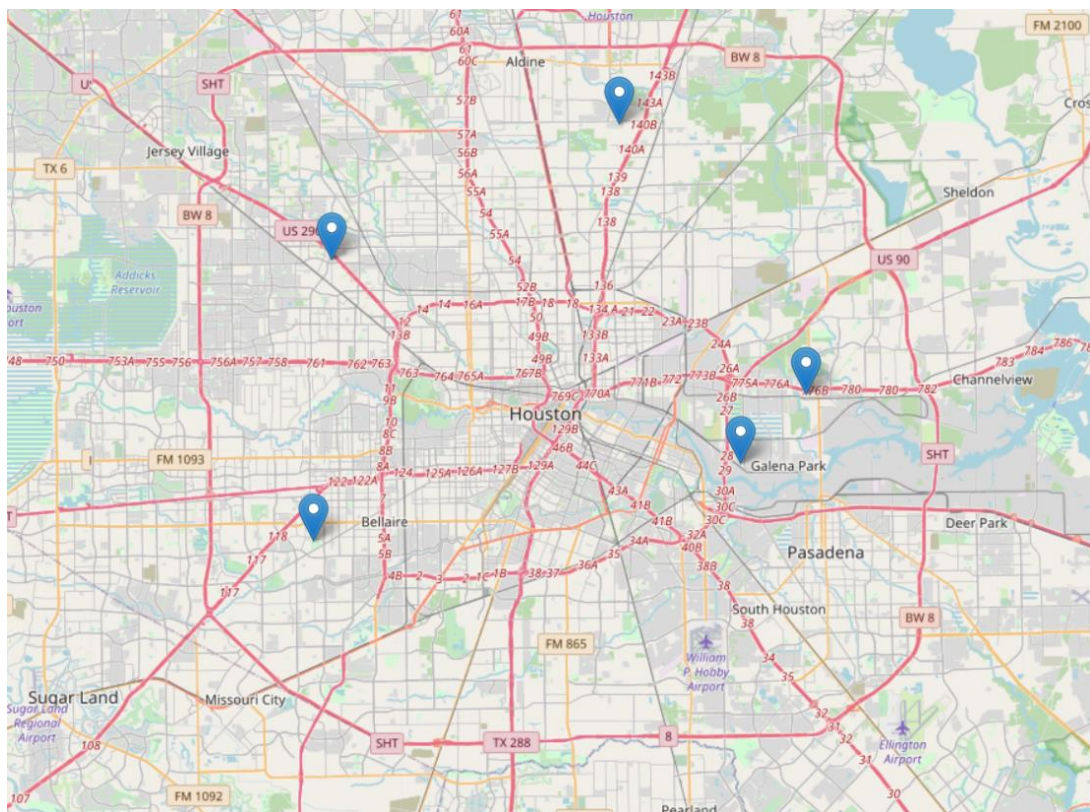


Figure 2.3: Long-running ambient air quality sensor locations in Houston, TX

2.3 Results and Discussion

In general, fuel sales increased during the period 1990 to 2019, with higher relative growth rates for diesel fuel than for gasoline, and higher growth rates in the US than in California (Figure 2.4). Over a nearly thirty-year period, diesel fuel sales have about doubled in the US and increased by nearly 70% in California. The corresponding increases in gasoline sales were 29% nationally and 17% in California from 1990-2019, before dropping during the Covid-19 pandemic in 2020. The effects of the Great Recession are clearly apparent in the diesel fuel sales data, which shows a significant drop-off between 2007 and 2009. After a decade of stable or slightly declining gasoline sales since 2002, a gradual increase in gasoline sales is observed from 2003-2019. Increasing fuel economy could help slow increasing gasoline fuel sales trends, however, fuel economy improvements have been slow since 1990, increasing only slightly for new light-duty vehicles from 21.2 mpg in 1990 to 25.5 mpg in 2019.⁸¹ For both gasoline and diesel, increases in fuel sales must be offset with a more rapid decrease in NO_x emission factors to deliver decreases in NO_x emissions. This objective is particularly challenging to achieve for heavy-duty diesel engines, where the rate of growth in fuel sales has been much higher than for gasoline.

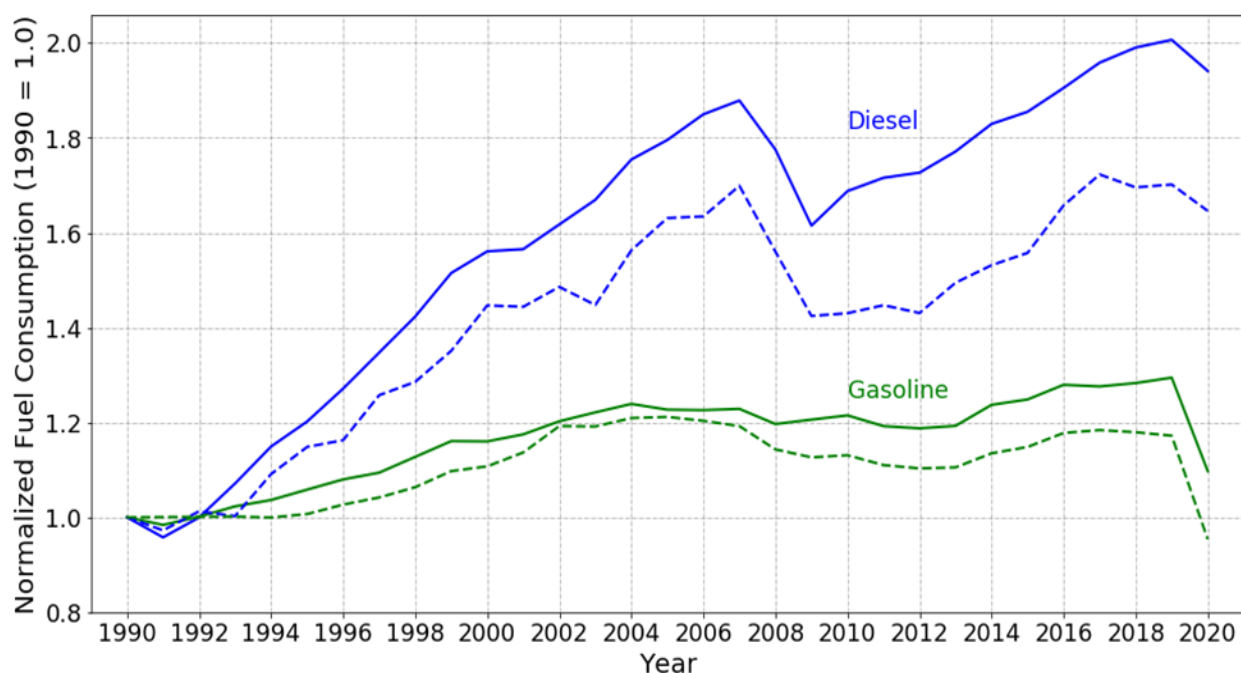


Figure 2.4: Trends in National (solid lines) and California (dashed lines) taxable sales of gasoline and diesel fuel intended for on-road use, for the period 1990-2020. Fuel sales for all years shown are normalized to corresponding 1990 values.

NO_x emission factors for light-duty gasoline vehicles have decreased dramatically: since 1990, emission factors have decreased from 14-18 to 2-3 g kg⁻¹ (Figure 2.5). The US trend shows a larger range of uncertainty due to the wider spread of measured values prior to 2010. The absolute rate of decrease in emission factors has slowed since 2005, which led us to use polynomial rather than linear fits to data points from on-road measurement studies.

Light/medium-duty diesel emission factors remain significantly higher than light-duty gasoline, but follow a different trend than heavy-duty diesel, with an estimated decrease from 30 to 19 g kg⁻¹ since 1990 (Figure 2.5). This decline has been slow overall, and the results indicate that light/medium-duty diesel emission factors may now be the highest of any category in recent years when compared to the rapidly declining emission factors in light-duty gasoline and heavy-duty diesel vehicles. While data from chassis dynamometer studies were not directly included in this analysis, dynamometer testing of light-duty gasoline of Kansas City vehicle fleets in 2004 and 2005 were calculated for comparison purposes and found to be similar to the light-duty gasoline fleet averages from the 2003 and 2005 Denver remote sensing campaigns.⁸²

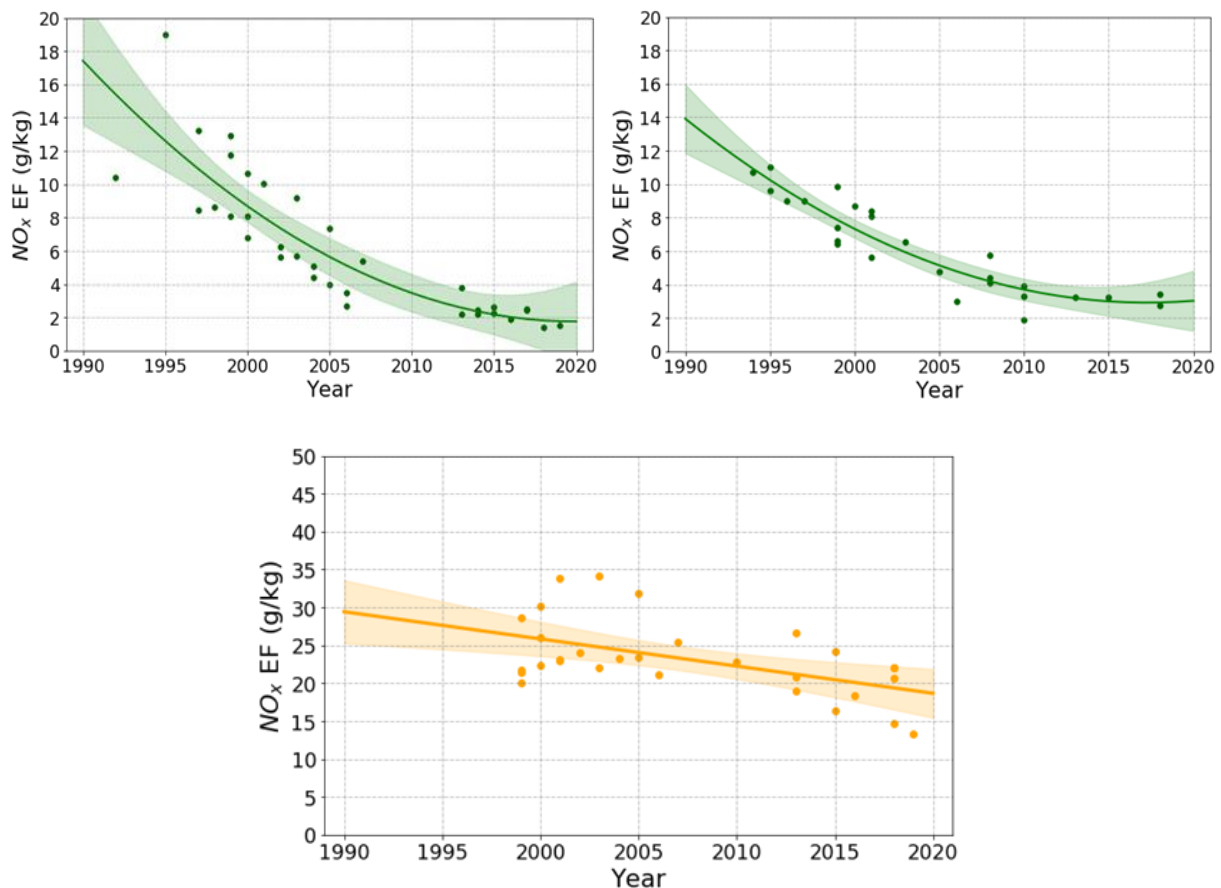


Figure 2.5: Trends in measured fleet-average NO_x emission factors for US light-duty gasoline vehicles (i.e., non-California locations; top left panel), California light-duty gasoline vehicles (top right panel), and combined US and California light/medium-duty diesel vehicles (bottom panel) for calendar years 1990-2020. Emission factors are expressed per unit mass of fuel burned. The regression-derived shaded bands shown in these plots indicate 95% confidence intervals (Table 2.3).

Table 2.3: Regression equation coefficients for emission factor trends with 95% confidence intervals

	Equation	Coefficients
US Gasoline	$A + Bx + Cx^2$	$A = 71708 \pm 6.15e+04$ $B = -71.001 \pm 61.3$ $C = 0.017576 \pm 0.0153$
CA Gasoline	$A + Bx + Cx^2$	$A = 60276 \pm 3.73e+04$ $B = -59.758 \pm 37.1$ $C = 0.014812 \pm 0.00926$
CA Diesel (Post-2004)	$A + Bx$	$A = 3715.8 \pm 1.18e+03$ $B = -1.8353 \pm 0.585$
Light Duty Diesel	$A + Bx$	$A = 743.71 \pm 446$ $B = -0.35894 \pm 0.222$

The fleet-average NO_x emission factor for the legacy fleet of older heavy-duty diesel trucks was assumed to remain constant at $38 \pm 5 \text{ g kg}^{-1}$ up until calendar year 2004. This choice is consistent with the emission factor measurements in the early-2000s (Figure 2.6) and results in increased diesel emission estimates in the late-1990s to mid-2000s. As a point of comparison to laboratory testing on the heavy-duty side for consistency, a chassis dynamometer study in 2015 measured similar fuel-normalized emission factors for SCR-equipped trucks to an on-road study under the same driving conditions, both capturing high and low SCR activity periods with comparable fuel-normalized emission factors.²⁹ The downward trend in NO_x emission factors since the calendar year 2004 is well-supported by on-road measurements made at multiple sites in California, represented as solid circles in Figure 2.6. The trendline for California is expected to show a steeper decline than for the US because California has required accelerated replacement of older diesel engines with newer equipment that meet the heavy-duty engine standards for NO_x and PM. In the absence of relevant on-road measurements outside of California, the dashed line shown in Figure 2.6 estimates the downward trend in NO_x emission factors for the national heavy-duty diesel truck fleet. Additional measurements of diesel truck emissions at non-California sites are recommended to augment the existing record of on-road emissions data shown and provide a clearer picture of national trends.

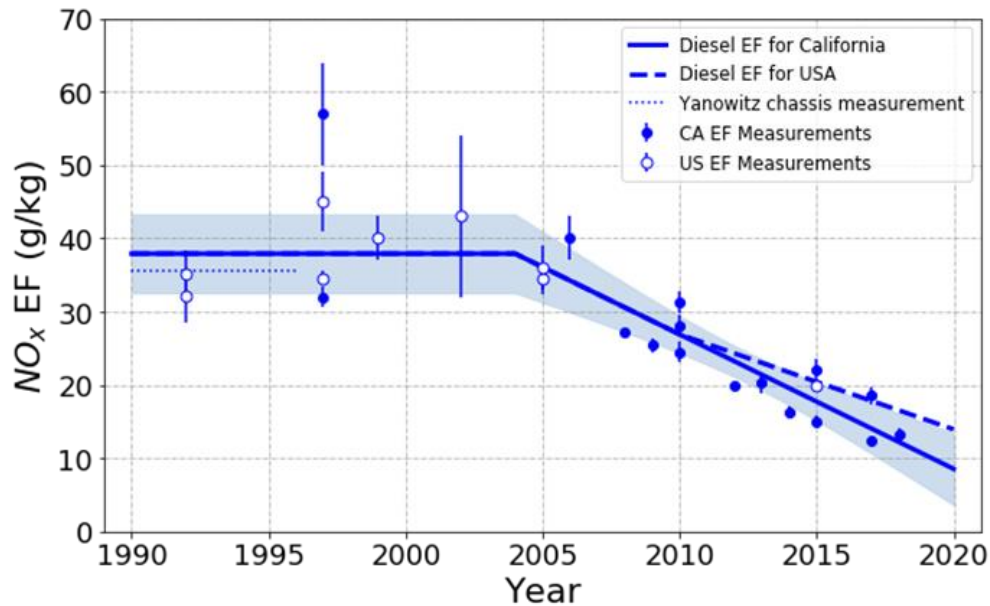


Figure 2.6: NO_x emission factor trends for on-road heavy-duty diesel engines for calendar years 1990-2020. Emission factors are expressed per unit mass of diesel fuel burned. NO_x emission factors for California (solid line and filled circles) are the same as for the US national fleet (dashed line and open circles) up until the calendar year 2010. After 2010, the lines diverge due to the accelerated replacement of the legacy fleet of older trucks in California (see text). The dotted line at the left represents the results of chassis dynamometer measurements of emissions from older diesel trucks.²⁰ The shaded bands shown indicate 95% confidence intervals based on regression values (Table 2.3).

Despite the significant increase in fuel consumption between 1990 and 2019, there has been a large decrease in vehicle-related NO_x emissions both for the US as a whole and for California (Figure 2.7 and Table 2.4). While NO_x emissions from gasoline engines have been decreasing for the entire period, diesel NO_x emissions exhibit an increasing trend prior to 2004, when fuel use was increasing and NO_x emission factors were not changing. After 2004, diesel NO_x emissions have been decreasing due to decreases in emission factors that have been sufficient to outpace growth in fuel sales and diesel truck travel. An exceptionally rapid decrease in emissions is observed during the period 2007-2009, resulting from a period of decreased diesel fuel sales during the Great Recession. The diesel-source NO_x emissions decrease is especially notable in California, where total diesel emissions over the last ten years have decreased to levels that are comparable to total gasoline emissions. This is not estimated to be true in the US, where diesel remains a higher fraction of the total. The post-2010 rate of decrease in diesel NO_x emissions was approximately 32% in the US and 48% in California, further emphasizing the influence of the rollout of SCR-equipped trucks. From 1990-2019, the overall change in total NO_x emissions from on-road vehicles was similar in the US and California, with decreases of $69 \pm 19\%$ and $71 \pm 15\%$ in the US and California, respectively.

EMFAC gasoline emission estimates are largely within our 95% confidence interval and follow the same trend as the fuel-based inventory in most years. With the additional consideration for light/medium-duty diesel in the fuel-based analysis, EMFAC predicts higher

diesel emissions for most of the 2000s but follows a similar rate of decrease and closes the gap with the fuel-based inventory in the mid-2010s. The MOVES model-derived emission trends differ significantly from the fuel-based inventory: overall NO_x emissions estimates from the fuel-based inventory are decreasing less rapidly than predicted by the MOVES model. MOVES predicts higher diesel emissions in the earlier years with a steeper rate of decrease, as well as higher gasoline emissions for the entire period of analysis. The higher MOVES estimates for gasoline could be in part due to higher estimates of cold start-related emissions from gasoline engines compared to the fuel-based inventory, though this issue does not affect the diesel emissions comparisons and has a reduced impact when evaluating total on-road emissions since the diesel-source NO_x contribution is dominant. We expect the rate of decrease in diesel NO_x emissions to be slower outside of California, where accelerated replacement of older trucks is not required, whereas MOVES predicts a more rapid rate of decrease in NO_x emissions compared to both EMFAC and the present fuel-based inventory. MOVES also fails to capture the recession-related decrease in diesel fuel sales that occurred between 2007 and 2009 and predicts that gasoline and diesel emissions have converged much more than the fuel-based inventory suggests.

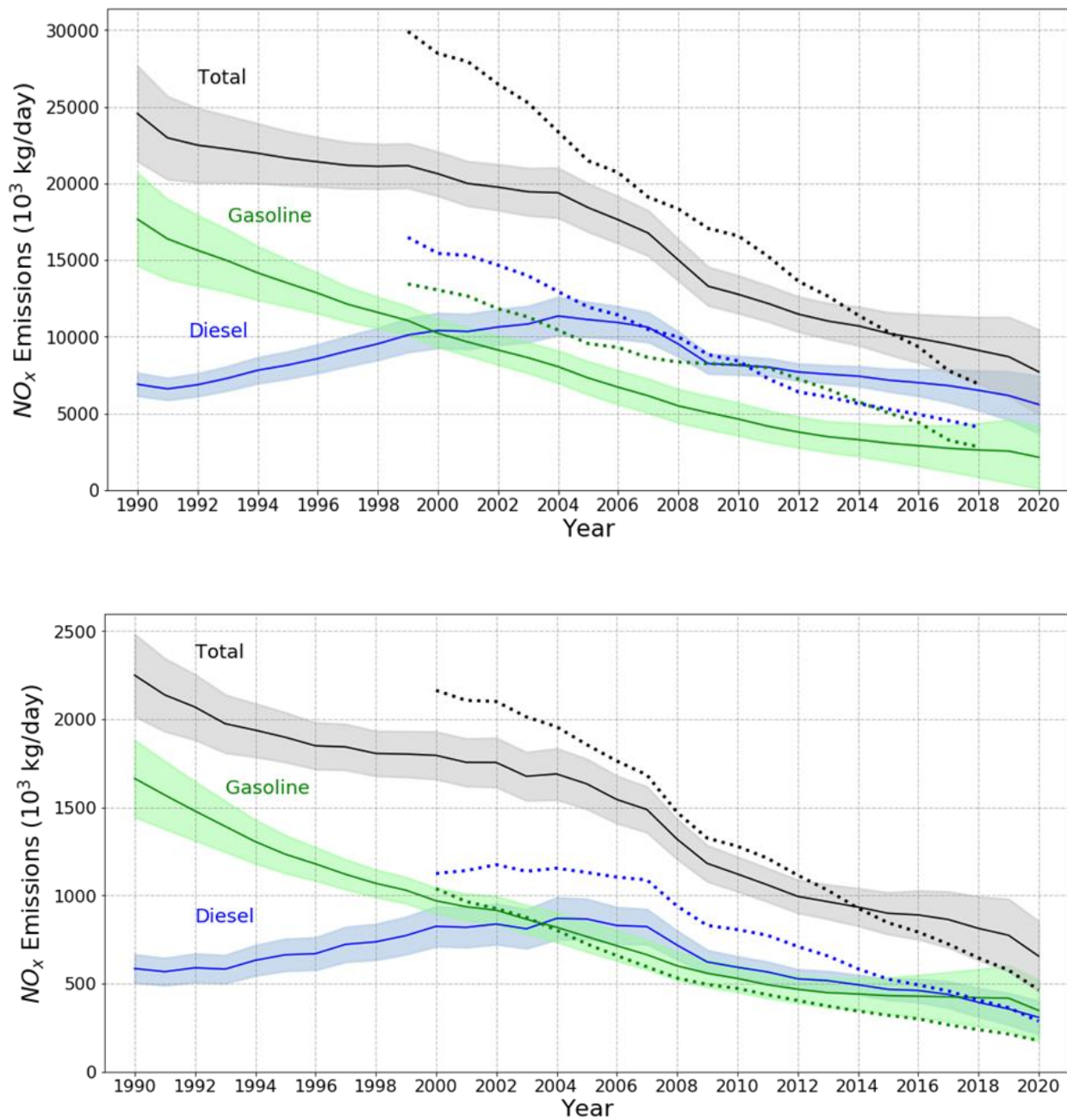


Figure 2.7: National (top) and California (bottom) trends in NO_x emissions from on-road vehicles for the period 1990-2020. Solid lines and associated shaded bands denote best estimates and 95% confidence intervals for the fuel-based emission inventory. For comparison purposes, dotted lines indicate emission inventory estimates from the MOVES and EMFAC models for the U.S. and California, respectively. Excess emissions associated with cold engine starting have been included in gasoline emission inventory estimates shown here (see text).

Table 2.4: National and California fuel-based on-road NO_x emission estimates

Type	Year	Diesel Emissions (tpd)	Diesel Error (tpd)	Gasoline Emissions (tpd)	Gasoline Error (tpd)	Total Emissions (tpd)	Total Error (tpd)
California	1990	585	80	1664	219	2250	233
	1991	568	78	1570	192	2138	207
	1992	590	81	1480	167	2070	186
	1993	582	80	1393	145	1975	165
	1994	632	87	1306	124	1938	152
	1995	664	91	1234	108	1898	141
	1996	670	92	1179	95	1850	132
	1997	723	99	1120	85	1843	130
	1998	737	101	1069	78	1806	128
	1999	773	106	1030	75	1803	130
	2000	825	114	970	73	1795	135
	2001	819	113	936	75	1756	135
	2002	838	115	916	80	1755	140
	2003	811	111	866	81	1677	137
	2004	870	120	820	84	1690	146
	2005	867	114	767	85	1634	142
	2006	830	104	715	85	1545	134
	2007	824	99	664	84	1488	130
	2008	719	82	601	80	1320	114
	2009	623	67	559	78	1182	103
	2010	593	62	529	77	1122	99
	2011	566	58	494	76	1060	95
	2012	527	54	468	77	995	94
	2013	517	54	448	81	965	98
	2014	494	56	441	91	935	107
	2015	468	59	432	103	899	119
	2016	461	66	429	121	890	138
	2017	439	75	425	141	864	159
	2018	394	81	420	162	814	181
	2019	357	89	418	186	775	206
	2020	308	95	348	174	656	198
National	1990	6892	744	17663	3024	24555	3114
	1991	6592	711	16381	2608	22973	2703
	1992	6861	740	15633	2306	22494	2422
	1993	7288	791	14952	2031	22239	2179
	1994	7806	846	14162	1758	21968	1951
	1995	8147	883	13495	1524	21642	1761
	1996	8566	932	12841	1317	21406	1613

	1997	9067	985	12110	1137	21178	1505
	1998	9530	1039	11580	1019	21110	1455
	1999	10108	1104	11051	947	21159	1454
	2000	10403	1136	10224	897	20627	1447
	2001	10339	1132	9650	903	19989	1448
	2002	10627	1160	9128	948	19755	1498
	2003	10825	1180	8627	1001	19452	1548
	2004	11347	1241	8041	1057	19388	1631
	2005	11112	1147	7307	1082	18419	1577
	2006	10921	1059	6704	1106	17625	1532
	2007	10614	965	6142	1121	16756	1479
	2008	9518	802	5484	1090	15002	1353
	2009	8245	645	5042	1086	13287	1263
	2010	8136	604	4623	1072	12759	1231
	2011	8009	572	4148	1029	12157	1177
	2012	7692	555	3773	1007	11465	1150
	2013	7548	581	3463	1012	11012	1166
	2014	7421	646	3278	1081	10699	1259
	2015	7161	731	3048	1167	10208	1377
	2016	6998	865	2884	1327	9881	1584
	2017	6800	1060	2718	1510	9518	1845
	2018	6497	1286	2601	1758	9099	2178
	2019	6154	1568	2536	2065	8690	2593
	2020	5571	1869	2130	2036	7702	2764

*All emission values in metric tons/day

To evaluate fuel-based emission inventory trends, we examined corresponding trends in measured ambient NO_x concentrations in Los Angeles and Houston (Figure 2.8 and Table 2.5). These cities have been the site of several field campaigns measuring NO_x in urban spaces, including a campaign in the Los Angeles area in 2010 and a campaign in the Houston-Galveston-Brazoria area in 2013.^{35,83} Decreases in ambient NO_x concentrations, measured on weekday mornings when traffic-related emissions dominate, correspond relatively well with downward trends in the fuel-based inventories. The decreases in total on-road NO_x emissions and ambient NO_x concentrations are 73% and 65% in Los Angeles and 64% and 48% in Houston, respectively. An implication of these comparisons is that if the rest of the US also had programs like California's to accelerate the replacement of older heavy-duty engines, the diesel NO_x source could be better controlled nationwide as well. Note that compared to state and national trends shown in Figure 2.7, the relative importance of the diesel NO_x source is generally equivalent to or lower than the importance of the gasoline NO_x source within urban areas as shown in Figure 2.8. This is expected because diesel truck traffic includes a higher proportion of travel on the highway network, including long-haul trips through less populated regions.

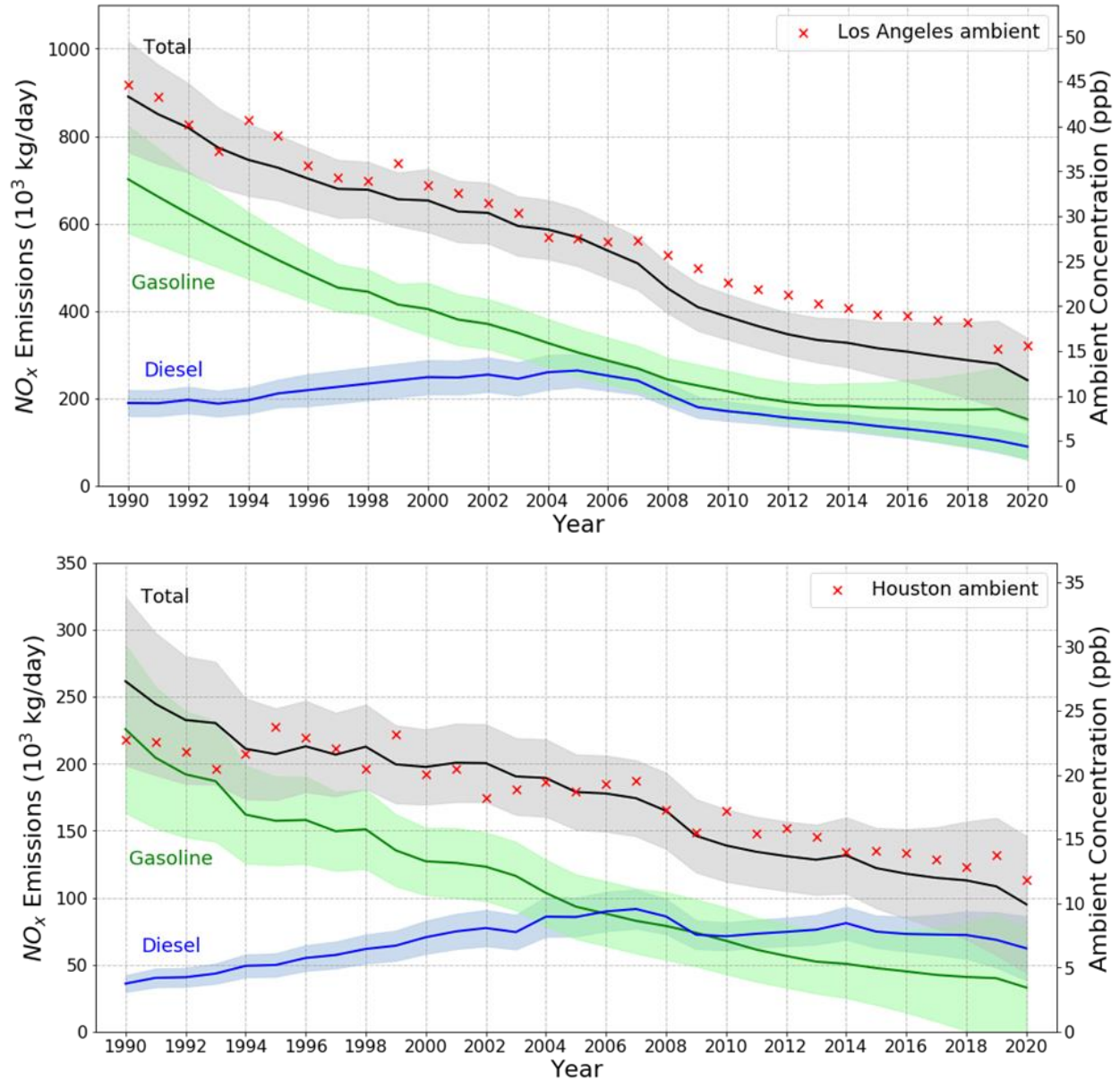


Figure 2.8: Trends in ambient concentrations and on-road vehicle emissions of NO_x for Los Angeles, California (top) and Houston, Texas (bottom). The red markers represent annual averages of ambient NO_x concentrations on weekday mornings at multiple long-running measurement sites in each city.

Table 2.5: Houston and Los Angeles area fuel-based on-road NO_x emission estimates and annual weekday morning averaged ambient NO_x data.

City	Year	Gasoline Emissions	Gasoline Error	Diesel Emissions	Diesel Error	Total Emissions	Total Error	Ambient NO _x (ppb)
Houston	1990	226	63	36	6	262	63	64
	1991	205	53	40	7	245	53	63
	1992	192	47	41	7	233	48	62
	1993	187	45	43	8	230	46	58
	1994	162	37	49	9	211	38	55
	1995	157	33	50	9	207	34	58
	1996	158	33	55	10	213	34	60
	1997	150	30	57	10	207	31	57
	1998	151	30	62	11	213	31	52
	1999	135	27	64	11	200	29	59
	2000	127	25	70	12	198	28	46
	2001	126	26	75	13	201	29	56
	2002	123	26	77	14	200	29	44
	2003	116	26	74	13	191	29	47
	2004	104	25	86	15	189	29	45
	2005	93	24	86	14	179	28	47
	2006	88	24	90	15	178	28	46
	2007	83	24	92	15	174	28	46
	2008	79	25	86	13	165	28	37
	2009	74	25	72	11	146	27	32
	2010	68	25	71	10	139	27	35
	2011	61	24	73	10	134	26	31
	2012	57	24	74	11	131	26	32
	2013	52	24	76	11	128	26	33
	2014	51	26	81	12	132	28	28
	2015	47	27	75	12	122	30	29
	2016	45	31	73	13	118	33	28
	2017	42	35	72	15	115	38	24
	2018	41	40	72	18	113	44	25
	2019	40	47	69	21	108	51	26
	2020	33	45	62	24	95	51	21
Los Angeles	1990	701	123	189	30	890	127	-
	1991	661	110	189	30	850	114	131
	1992	623	97	196	30	819	102	127
	1993	586	86	188	30	774	91	-
	1994	550	76	195	30	746	82	108
	1995	516	68	211	32	728	75	97

	1996	484	61	219	37	703	71	91
	1997	453	55	226	37	679	66	90
	1998	444	51	234	38	677	64	93
	1999	414	48	241	39	655	61	104
	2000	404	61	248	39	653	72	94
	2001	380	58	247	39	628	70	90
	2002	370	57	254	39	624	69	87
	2003	350	56	245	38	594	68	86
	2004	327	55	260	39	586	68	79
	2005	305	54	263	38	568	66	78
	2006	286	53	252	34	538	63	79
	2007	268	52	240	31	509	61	73
	2008	243	50	209	27	451	57	68
	2009	229	48	180	24	409	54	60
	2010	216	47	170	22	386	52	53
	2011	201	46	164	21	365	50	51
	2012	191	46	155	20	346	50	48
	2013	184	47	149	19	333	51	44
	2014	183	52	144	20	327	56	41
	2015	178	57	136	20	314	61	39
	2016	177	65	130	21	307	68	37
	2017	174	73	122	23	296	77	35
	2018	174	83	113	25	287	87	35
	2019	175	95	103	27	279	99	31
	2020	152	92	89	29	241	97	27

*All emission values in metric tons/day

While sales of both gasoline and diesel fuel have increased since 1990, the implementation of NO_x emission control technologies has lowered emission factors sufficiently to result in an overall decline in NO_x emissions from on-road vehicles over the same period. The downward trend in emissions strengthened noticeably starting around 2004 when changes in diesel NO_x emissions began to reinforce rather than offset progress in controlling NO_x emissions from gasoline engines. While the underlying reasons are not clear, there is a need for further examination into why some emission model estimates are not in line with fuel-based inventories, satellite studies, and field campaigns. California is well on its way to replacing all pre-2010 heavy-duty diesel truck engines by 2023, and the US national truck fleet will likely continue to lag in making progress in controlling NO_x emissions by allowing older pre-2010 engines to remain in the fleet.

For both light-duty gasoline and heavy-duty diesel vehicles, there are diminishing returns as emission factors begin to approach small values, meaning that future efforts to reduce NO_x emission factors for these vehicles may have limited impacts on NO_x emissions. However, evaluation of light/medium-duty diesel emission trends suggests room for improvement given

their high NO_x emission factors compared to more heavily regulated heavy-duty diesel vehicles. At present, downward trends in NO_x emissions are likely to flatten out, and emissions may start rising again if increases in fuel sales outpace future decreases in emission factors. Other approaches such as electrification of the vehicle fleet could be helpful going forward in reducing not only CO₂ but also NO_x emissions. Also, as NO_x emission factors from on-road engines have decreased substantially, other off-road mobile sources^{84,85} will play a larger role in contributing to NO_x emissions and ozone formation.

The trend in NO_x emissions from on-road and other sources has implications for ozone levels, especially in urban areas. Many US cities do not meet the current ozone air quality standard of 70 ppb,⁸⁶ and in general, cities have moved towards increasingly NO_x-sensitive chemical regimes, meaning that controlling NO_x will be more important than controlling VOC emissions.⁵⁶ Ozone levels are now decreasing only slowly over time, and there is an unhelpful increasing trend in background levels of tropospheric ozone in the Northern Hemisphere as a whole.^{87,88}

Chapter 3: Improved Spatial Resolution in Modeling of Nitrogen Oxide Concentrations in the Los Angeles Basin

This chapter is reproduced and adapted from:

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The emission inventory used in this study is available at the following URL: <https://csl.noaa.gov/groups/csl7/measurements/2021sunvex/emissions/>.

3.1 Introduction

Nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) are highly reactive air pollutants that are produced primarily from the combustion of fossil fuels. Exposure to ground-level NO_2 is associated with increased mortality due to respiratory and cardiovascular disease, and NO_x is a major precursor to tropospheric ozone (O_3), which has negative impacts on human health and the environment.^{6,7,89,90} Nitrogen oxides also react in the atmosphere to create acid rain and particulate matter in the form of aerosol nitrates.⁶ To manage the human health and environmental impacts of NO_x , an improved understanding of NO_x emission sources and the distribution of NO_x in high-population areas is needed.

In the United States, the development of emissions control technologies has been driven by laws and regulations aimed at reducing air pollution, with major changes starting in 1963 with the Clean Air Act and its subsequent amendments. The primary technologies to reduce NO_x emissions from vehicles involve devices that convert NO_x emissions into water and N_2 . The widespread adoption of catalytic converters has significantly decreased NO_x emissions from gasoline vehicles since the 1990s. As a result, heavy-duty diesel trucks became the largest mobile source of NO_x emissions as light-duty gasoline emissions declined and diesel fuel sales continued to grow^{18,19,91}; these trends were reinforced due to ineffective diesel NO_x emissions controls in the 1990s.²⁰ Selective catalytic reduction (SCR) systems were introduced on new heavy-duty diesel engines starting in 2010. SCR systems use a urea solution to convert NO_x to N_2 and have been shown to reduce in-use NO_x emissions from heavy-duty diesel engines by >75%.^{24–29} However, the effectiveness of these systems in reducing NO_x emissions under real-world driving conditions can be impaired at times due to factors such as extended idling and low exhaust temperature leading to inactivation of the emission control system.^{26,29}

The extent to which the adoption of more effective NO_x control technology has affected emission trends in the US remains unclear. Field campaigns, satellite-based inventories, and chemical transport models have produced results that highlight uncertainties in NO_x emission inventories, including how much NO_x is being emitted and the extent to which NO_x emissions have decreased in recent years. Estimates of NO_x based on the MOtor Vehicle Emission

Simulator (MOVES) emission model overestimated NO_x when compared to field campaigns in the Baltimore-Washington area and in the southeast US.^{44,45,92} The MOVES model also estimated larger decreases in NO_x emissions than NO₂ satellite retrievals suggest.⁴⁶ These differences are coupled with the fact that while precursors emissions to ozone have decreased over the past several decades, ozone concentrations in urban areas have declined relatively slowly and background ozone in the northern hemisphere has been slowly increasing.^{87,88}

Many urban areas in North America are transitioning towards NO_x-limited ozone formation regimes, implying that NO_x emission reductions are increasingly necessary to reduce ozone.^{56,93} The Los Angeles area is one of the largest and most populated urban areas in the United States, with air pollution problems that are linked to local topography, high volumes of vehicle traffic, and two major ports that handle nearly 30% of all imports and exports over the water in the US.³⁴ Ozone concentrations in the region remain high despite major reductions in ozone precursors and as a result, the South Coast Air Basin (SOCAB) was designated “extreme nonattainment” for ozone in 2018.^{91,94}

Chemical transport models (CTMs) have been a useful tool for understanding source contributions, supporting air quality planning, and informing policy decisions. However, in order to maintain reasonable computational time and costs, most modeling studies focused on NO_x chemistry remain at a spatial resolution too coarse to see individual roadway effects. While these setups have generally been able to answer questions about the distribution and sources of NO_x on state-wide and continental levels, they have generally not been useful for resolving sharp near-source gradients, especially near major roadways. At higher spatial resolution, there are several additional benefits including being able to model the local effects of highways, look at neighborhood-scale differences in human health exposure, and validate models with higher-resolution satellite retrievals. While the current highest resolution satellite measurements for NO₂ from the Tropospheric Monitoring Instrument (TROPOMI) include reprocessed data at a resolution of 3.5 km x 5.5 km⁴⁷, new instruments such as the Tropospheric Emissions: Monitoring of Pollution (TEMPO) instrument will provide even higher resolution data on pollutant concentrations in the future.⁴⁹ Moving to a high-resolution inventory is in line with recent improvements in satellite spatial resolution and the increasing need to examine how changes to emissions will affect the exposure of distinct communities to air pollutants.

Fuel-based inventory methods provide a complementary perspective to emission model predictions and trends inferred from satellite data, providing critical insights into how emission control technologies have impacted NO_x concentrations in heavily trafficked urban areas. To gain a better representation of the temporal and spatial patterns of NO_x, this research develops and evaluates a 1.3 km x 1.3 km high-resolution fuel-based inventory for Los Angeles, CA. This study evaluates this inventory as input in the Weather Research and Forecasting with Chemistry (WRF-Chem) model by comparing aircraft measurements taken during the June 2021 RECAP-CA campaign and vertically-integrated tropospheric NO₂ concentrations from the Sentinel-5P/TROPOMI satellite. The resulting spatial and temporal agreement with observational data allows us to understand the individual contributions of NO_x emission sectors in a major urban area.

3.2 Methods

3.2.1 Atmospheric Model and Study Domain

WRF-Chem (version 4.2.2⁵⁰) is used to predict meteorological and air quality-related variables for all of June 2021, using the last three days of May as a model spin-up period. The model was applied over two nested domains: (1) all of California and Nevada at 4 km horizontal resolution, and (2) southern California at an unusually fine horizontal resolution of 1.3 km. The model includes 50 vertical levels up to 50 hPa. Initial and boundary conditions for chemical species tracked in the model are from a parent 12 km resolution continental US simulation that used a similar model setup and covered the same period. Initial and boundary conditions for meteorological variables are from the 3 km horizontal resolution High-Resolution Rapid Refresh (HRRR) model, which features hourly assimilation of meteorological variables such as wind and temperature from in-flight commercial aircraft, as well as ground-based radar reflectivity observations of precipitation.⁹⁵ Several meteorological setups were tested and compared to RECAP-CA aircraft wind measurements. Using HRRR gave better model performance for wind speed and direction relative to setups using the North American Mesoscale (NAM) and Rapid Refresh (RAP) models. The chemical mechanism used in this study is a version of the Regional Atmospheric Chemistry Mechanism⁹⁶ with updates to account for key oxygenated volatile chemical product (VCP) emissions (RACM-ESRL-VCP) as specified in Coggon et al.^{96,97} Chemical vertical mixing is enhanced for low boundary layer height under polluted and fire conditions. Further details of the input data and WRF-Chem model parameterizations are included in Table 3.1.

Table 3.1: Summary of WRF-Chem model setup and input data

WRF-Chem model version	WRFv4.2.2
Horizontal resolution	D01: 4km x 4km over California and Nevada D02: 1.3km x 1.3km over southern California
Vertical resolution	50 vertical levels from the surface to 50 hPa
Initial/boundary conditions (meteorology)	High-Resolution Rapid Refresh (HRRR) at 3 km resolution ⁹⁸
Initial/Boundary conditions (chemistry)	12 km resolution WRF-Chem run using NAM meteorology and chemical boundary conditions from the Realtime Air Quality Modeling System (RAQMS)
Gas phase chemistry	RACM-ESRL-VCP ⁹⁷
Photolysis scheme	Madronich photolysis (TUV)
Anthropogenic emissions	Modified Fuel-based Inventory of Vehicle Emissions (FIVE) ⁹⁹ , Fuel-based oil and gas (FOG) inventory ¹⁰⁰ , Continuous Emission Monitoring Systems (CEMS) ¹⁰¹ , NEI 17 scaled to 2021 ⁵² , Copernicus Atmosphere Monitoring Service (CAMS) ¹⁰²
Biogenic emissions	Biogenic Emissions Inventory System (BEIS3.14) ¹⁰³ with updated isoprene and monoterpenes from the urban land cover type ^{35,104}
Additional parameterizations	Mellor-Yamada Nakanishi and Niino surface layer, Mellor-Yamada Nakanishi and Niino level 2.5 planetary boundary layer, Grell-Devenyi (GD) ensemble cumulus, Noah land surface model

3.2.2 Emission Inventory

Spatially- and temporally-resolved emissions from on-road vehicles in southern California were estimated and mapped using a modified version of the Fuel-based Inventory for Vehicle Emissions (FIVE).^{80,99} The methodology used to create the FIVE inventory and temporal scaling factors can be found in further detail in Harkins et al.⁹⁹ Emissions are distributed spatially using 2018 data from a national database of traffic counts from the Federal Highway Administration and are scaled to June 2021 using state-wide taxable fuel sales.^{60,105} Comparing the 2018 FHWA traffic counts to state-level traffic statistics, the traffic count data accounts for the majority of fuel use both for gasoline (68%) and diesel fuel (77%) in California.⁵⁹ Remaining fuel use is mostly on small local roads rather than major roadways, and can thus be adequately distributed using census block-level population data gridded to 1.3 km resolution.¹⁰⁶ Gridded gasoline and diesel fuel consumption estimates are combined with fuel-specific emission factors (g of pollutant emitted per kg of gasoline or diesel fuel burned) to calculate emissions. Emission factors for each vehicle type are determined based on regression analyses of measurements from roadside remote sensing and highway tunnel studies conducted in California.⁹¹

The major modifications to the FIVE inventory for this study include moving to 1.3km resolution and the reapportionment of 22% of total diesel fuel to a light-/medium-duty diesel vehicle category, separate from the existing gasoline and heavy-duty diesel vehicles categories. These light/medium-duty diesel vehicles now have higher fuel-based NO_x emission factors compared to heavy-duty diesel trucks.⁹¹ Reasons for higher emission factors in this category may vary due to the inclusion of a wide range of vehicle types, but contributing factors include slow fleet turnover of light-duty diesel engines and less deployment of advanced emission control technology (i.e., selective catalytic reduction for NO_x control) in comparison to heavy-duty diesel trucks. Heavy-duty diesel engines have been a high state-wide priority in efforts to accelerate the replacement of older (pre-2010) engines with newer and lower-emitting engines.⁷⁴ Diurnal and day-of-week variations in vehicular emissions are based on weigh-in-motion traffic count data, with separate temporal variation profiles for light- and heavy-duty vehicles.⁸⁰ We assume the spatial distribution of light and medium-duty diesel truck traffic matches that of light-duty gasoline vehicles and use the activity profile for heavy-duty trucks to specify temporal variations. In comparison to the existing FIVE inventory, this reapportionment of diesel fuel to light/medium-duty vehicles results in higher on-road NO_x emissions overall, especially on weekdays, and more emissions attributed to light-duty vehicle spatial patterns.

In addition to on-road vehicle emissions, the emission inventory used in this study includes fuel-based emissions from off-road engines used in agricultural and construction equipment.^{84,99,107} Also included were emissions from oil and natural gas production¹⁰⁰, power plants based on Continuous Emission Monitoring System data¹⁰¹, and other point and area sources from the 2017 National Emission Inventory scaled to 2021 based on activity factors.^{52,99} Oceangoing vessel emissions and emissions from all sources in Mexico were from the Copernicus Atmosphere Monitoring Service (CAMS) inventory.¹⁰² Biogenic emissions are based on the Biogenic Emissions Inventory System (BEIS) v3.14 model.^{103,108} Consistent with previous modeling work in Los Angeles³⁵, BEIS emissions for isoprene and monoterpenes from the urban land cover type were updated based on Scott and Benjamin.¹⁰⁴

3.2.3 Aircraft and Surface Monitor Data

Model predictions were compared to vertical profile NO_2 measured during aircraft flights over the Los Angeles basin that took place during the summer of 2021 as part of the RECAP-CA (Re-Evaluating the Chemistry of Air Pollutants in California) field campaign.^{109,110} Relevant flights occurred at midday hours on three weekend days (June 6, 12, and 19) and six weekdays (June 1, 4, 10, 11, 18, and 21). NO_x was measured at 5 Hz using a thermal dissociation laser-induced fluorescence (TD-LIF) instrument, and a detailed overview of the instrument can be found in Thorton et al. 2000 and Day et al. 2002.^{111,112} Instrument calibration details and methodology used during this campaign can be found in Zhu et al.¹¹⁰ Here, we report only data from the stacked racetrack patterns (see Figure 3.1) where the plane flew 4-6 different attitudes layer stacked on top of one another within the planetary boundary, which were designed to measure vertical concentration profiles. The aircraft data used here was split into flights that occurred closer to the coast (west/central LA) and at locations further inland (east basin) where temperatures were higher. Only altitude bins with greater than 5 observation points were used in the comparison. The aircraft data were matched with corresponding model predictions using the nearest neighbor method, averaging the aircraft data every 30 seconds and pulling comparison data for the nearest model grid point.

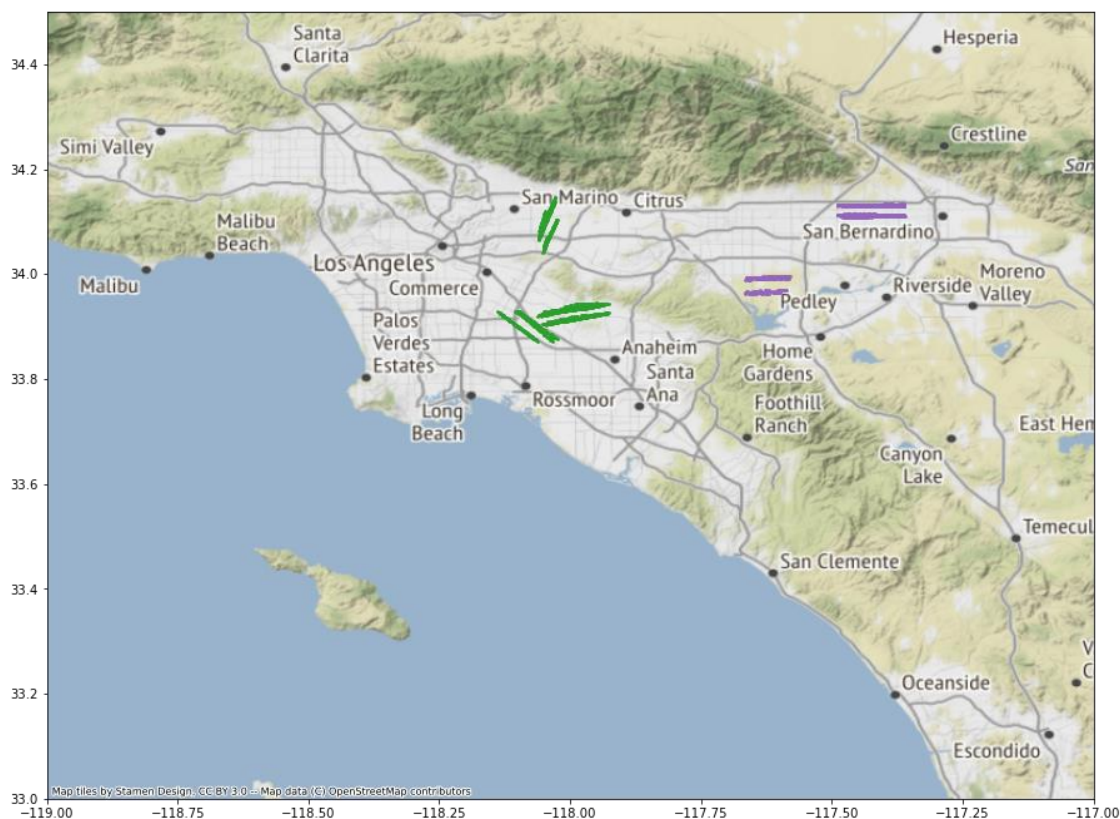


Figure 3.1: Flight paths used for vertical profile comparison. NO_2 measured in LA West (green) is separated from measurements taken in LA East (purple) to account for spatial differences in temperature.

In addition to verifying the vertical profiles against aircraft observations, the South Coast Air Quality Management District (AQMD) monitoring network was used for comparison to verify the diurnal patterns of NO₂ in the model.³⁵ The NO₂ data for 24 sites located in the South Coast air basin were averaged for each hour of the day on weekdays and weekends. The quantity NO₂* (NO₂* = NO₂ + PAN + alkyl nitrates + HONO + 2*N₂O₅) was used from the model for comparison to the measured NO₂ due to the conversion of other nitrogenous species in addition to NO₂ by the molybdenum converter installed within standard chemiluminescent NO_x analyzers.^{113,114}

3.2.4 Satellite Observations

TROPOMI satellite measurements are used in this study to evaluate tropospheric column-integrated model predictions of NO₂ concentrations. We use the latest NO₂ product reprocessed to a spatial resolution of 3.5 x 5.5 km² from Sentinel-5P Products Algorithm Laboratory (SP5-PAL) for June 2021.^{47,48} We used this reprocessed data product to create average June tropospheric NO₂ columns using methods described by Li et al., as well as separate weekday and weekend average tropospheric NO₂ columns for the study period.¹¹⁵ Pixels with cloud cover, snow or ice, or with otherwise problematic retrievals are filtered out to reduce uncertainties by using only data where the quality flag ≥ 0.75 .¹¹⁶ NO₂ vertical profiles require an assumption for the air mass factor (AMF), which may affect the ability to compare tropospheric NO₂ columns between the model and satellite data. In order to eliminate biases introduced by *a priori* profile assumptions, the NO₂ vertical profiles used in this study are calculated using averaging kernels, and the AMF is calculated from WRF-Chem, rather than using the AMF derived from the *a priori* TM5 Model.¹¹⁷ This approach has been shown to increase the satellite NO₂ concentrations over urban areas by 20% on average.¹¹⁵ Even accounting for these improvements, there is still an expectation of a low bias of -20% in TROPOMI tropospheric NO₂ columns over polluted cities in comparison to observations based on studies comparing to ground-based Pandora measurements.^{48,115,118}

3.3 Results and Discussion

Figure 1 shows the spatial distribution of NO_x emissions from on-road vehicles for the 1.3 km resolution southern California modeling domain. NO_x emissions are highest in the urban center, with lower emissions in sparsely populated rural and mountainous areas. This pattern follows from the emissions mapping methodology which relies on vehicle traffic counts that indicate high traffic densities within urban areas. Sharp spatial gradients in vehicle emissions near major highways are clearly apparent in the top panel of Figure 3.2, not only in rural areas but also within densely populated urban areas. While high NO_x emissions in and around downtown LA are mostly due to high volumes of light-duty vehicle traffic, elsewhere along major highways such as I-5 (running to the northwest of LA) and I-15 (running to the northeast of LA toward Las Vegas), the majority of the NO_x emissions are due to heavy-duty diesel trucks. Even though emissions from on-road vehicles decrease on weekends primarily due to the steep weekend decrease in diesel vehicle activity, vehicles overall are still the dominant source of NO_x emissions, responsible for 52% of NO_x on weekends versus 62% of NO_x on weekdays. The fraction of the total emissions from the on-road sector on weekdays can be seen in the bottom panel of Figure 3.2.

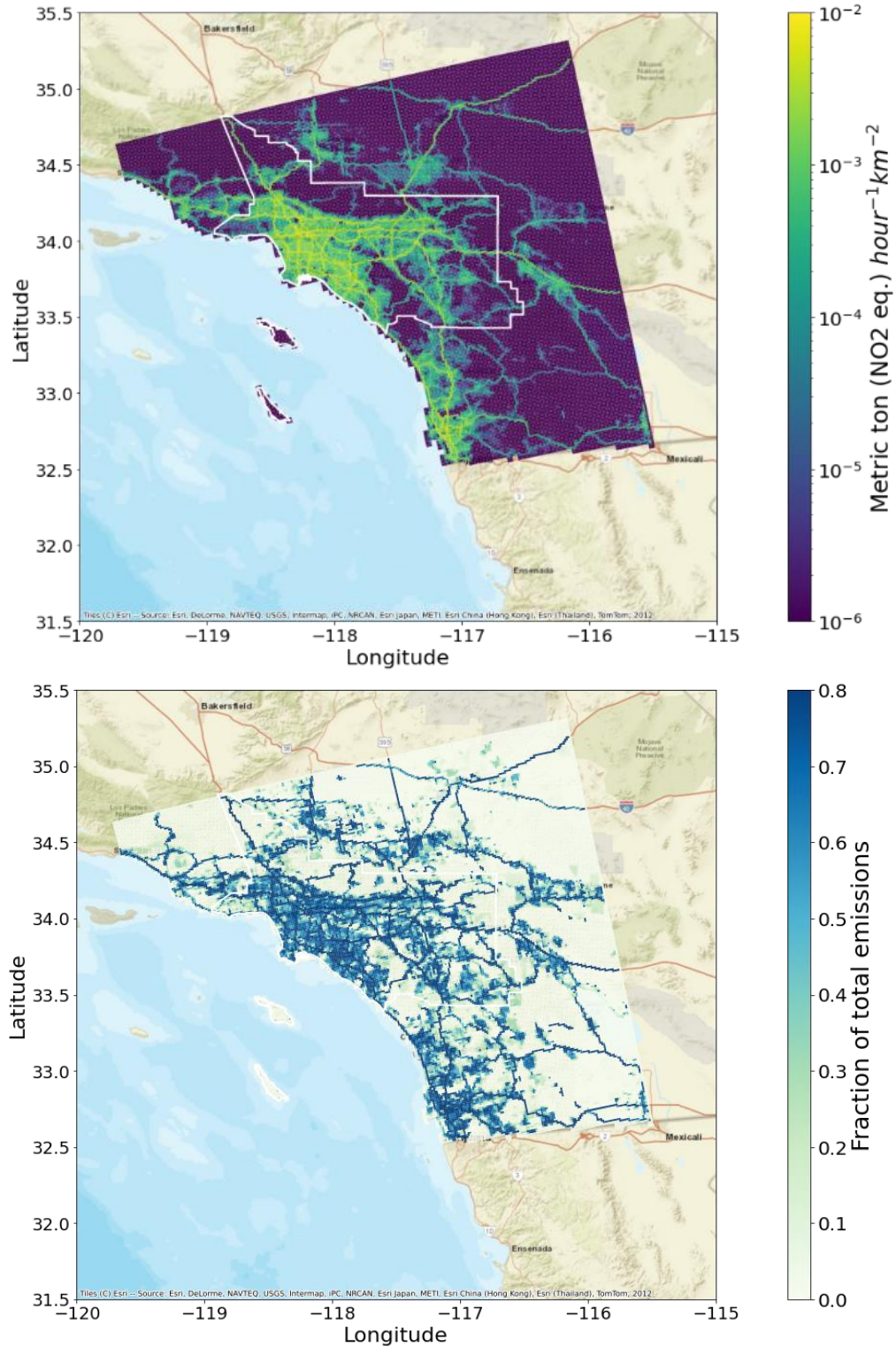


Figure 3.2. Spatial distribution of average 2021 weekday NO_x emission rates from on-road vehicles in southern California (top) and the fraction of total emissions from on-road (bottom), mapped at 1.3 km horizontal resolution. The South Coast Air Basin is outlined.

More details concerning NO_x emissions from on-road vehicles are presented in Table 3.2. We estimate that average daily NO_x emissions for the South Coast Air Basin were 198 ± 69 metric tons/day during our June 2021 study period, which is 1.6 ± 0.5 times the corresponding summer-season estimate from the California EMFAC model. Our estimates of NO_x emissions are comparable for heavy-duty diesel and higher for gasoline and light-/medium-duty diesel categories, with the largest relative difference (a ratio of 2.8 ± 0.6) for light- and medium-duty diesel vehicles. As shown in Table 3.2, on-road vehicle emissions are lower on weekends than on weekdays as expected, due mainly to decreased activity and emissions from diesel trucks. The overall decrease in vehicular NO_x emissions on weekends is about 100 metric tons per day, which is a 43% reduction relative to baseline weekday conditions. A potential cause of the lower emissions in the EMFAC model in comparison to the fuel-based inventory is in how emission factor trends are derived. The modified FIVE inventory used in this study uses on-road remote sensing data trends, resulting in emission factors that are higher overall and are decreasing more slowly in comparison to laboratory-measured emission factors used in other inventories. The large sample size of remote sensing data is better equipped to capture the effects of individual vehicles with ineffective or non-functioning emission control systems. While the remote sensing studies capture driving conditions that reflect typical vehicle activity patterns, they do not capture start-up and idling conditions, leaving room for actual average emission factors to be even higher. In the case of heavy-duty vehicles, selective catalytic reduction (SCR) systems do not operate effectively at low temperatures, leading to potential underestimates of mobile source emissions in urban areas when idealized SCR performance is assumed.

Table 3.2. Estimates of NO_x emissions from on-road vehicles in southern California^a

	vehicle category			
	gasoline	LD+MD diesel^b	HD diesel^b	On-road total
Fuel burned (t/day)	41763±2506	2295±298	6015±782	-
EF NO_x (g/kg)	2.0±0.6	20.2±3.4	9.2±4.6	-
Avg emissions (t/day)	87±27	49±10	62±32	198±69
WD^c emissions (t/day)	89	60	76	224
WE^c emissions (t/day)	82	20	26	127
EMFAC (t/day)	59	17	48	124
fuel-based/EMFAC^d	1.5±0.4	2.8±0.6	1.3±0.7	1.6±0.5

Notes:

^a Emission estimates for the South Coast Air Basin, including Los Angeles, Orange County, and portions of Riverside and San Bernardino Counties.

^b Diesel vehicle weight categories are light-, medium-, and heavy-duty (LD, MD, and HD).

^c Emissions for average weekday (WD) and weekend (WE) conditions.

^d Ratio of fuel-based emission inventory (weighted average of WD and WE values) from this study with corresponding estimates for summer 2021 from the most current version of the EMFAC model.⁷⁶

Figure 3.3 shows modeled and measured NO_2 concentrations as a function of altitude above sea level; values are binned within 100-meter intervals of altitude. The plots are separated into two regions, Western LA and Eastern LA, in order to eliminate spatial biases. Flight tracks showing the specific locations of these measurements are shown in Figure 3.1. Normalized mean biases for NO_2 are -19 and -8% for western and eastern portions of Los Angeles, respectively. Vertical profiles are in good agreement at most altitudes, especially below 600 m. In Eastern LA, the NO_2 concentrations are closer to the observations near the surface and diverge slightly with increasing altitude. In Western LA, we see that generally the model and observation NO_2 match very closely, especially between 250-600m. Despite slight underestimation in the model compared to the aircraft observations overall, these results indicate that the vertical representation of NO_2 in the model closely resembles the NO_2 measured during the field campaign during the same period. An accurate vertical profile in the model is critical to the calculation of tropospheric NO_2 columns from the satellite, as the AMF used in the calculation is revised to use the NO_2 vertical profile shape from the WRF-Chem simulation and averaging kernels.

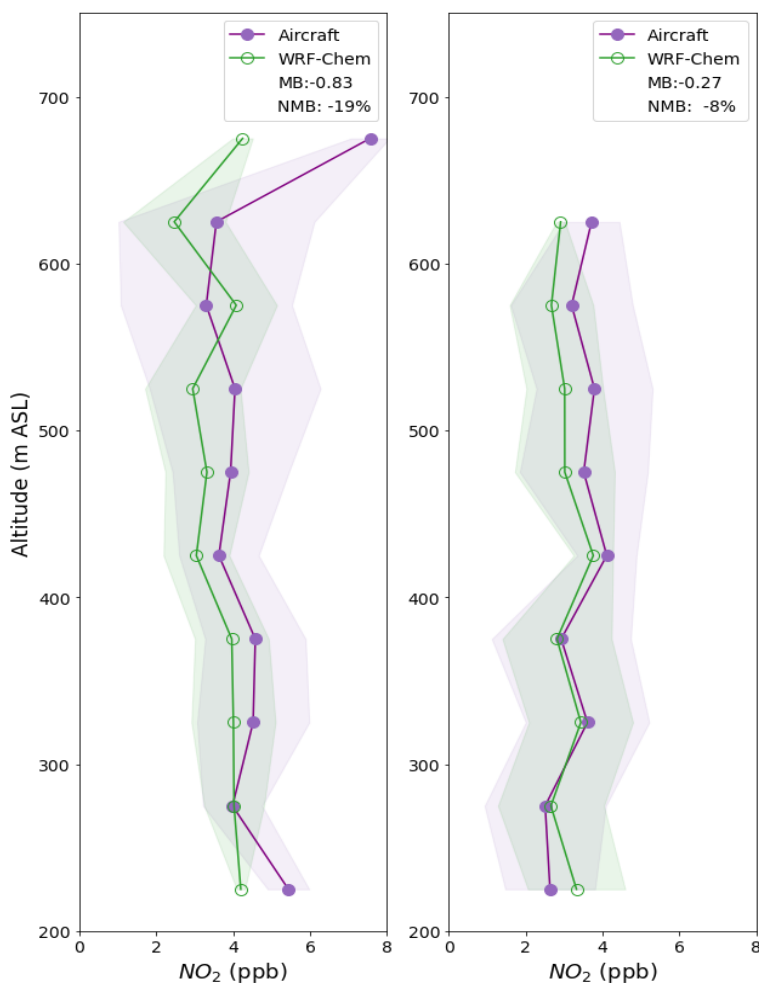


Figure 3.3: Vertical profiles of measured (RECAP flights) and modeled NO_2 concentrations over Western (left panel) and Eastern Los Angeles (right panel), binned over 100-meter intervals. Shaded bands indicate one standard deviation from the mean values.

The comparison of the diurnal patterns between the model and AQMD surface observations for the June 2021 study period is shown in Figure 3.4. This comparison suggests that the diurnal variations in modeled NO_2^* are consistent with observations, but there is some negative bias in modeled NO_2^* concentrations during nighttime hours on the weekends. The analysis performed at 4 km resolution yielded the same conclusions.

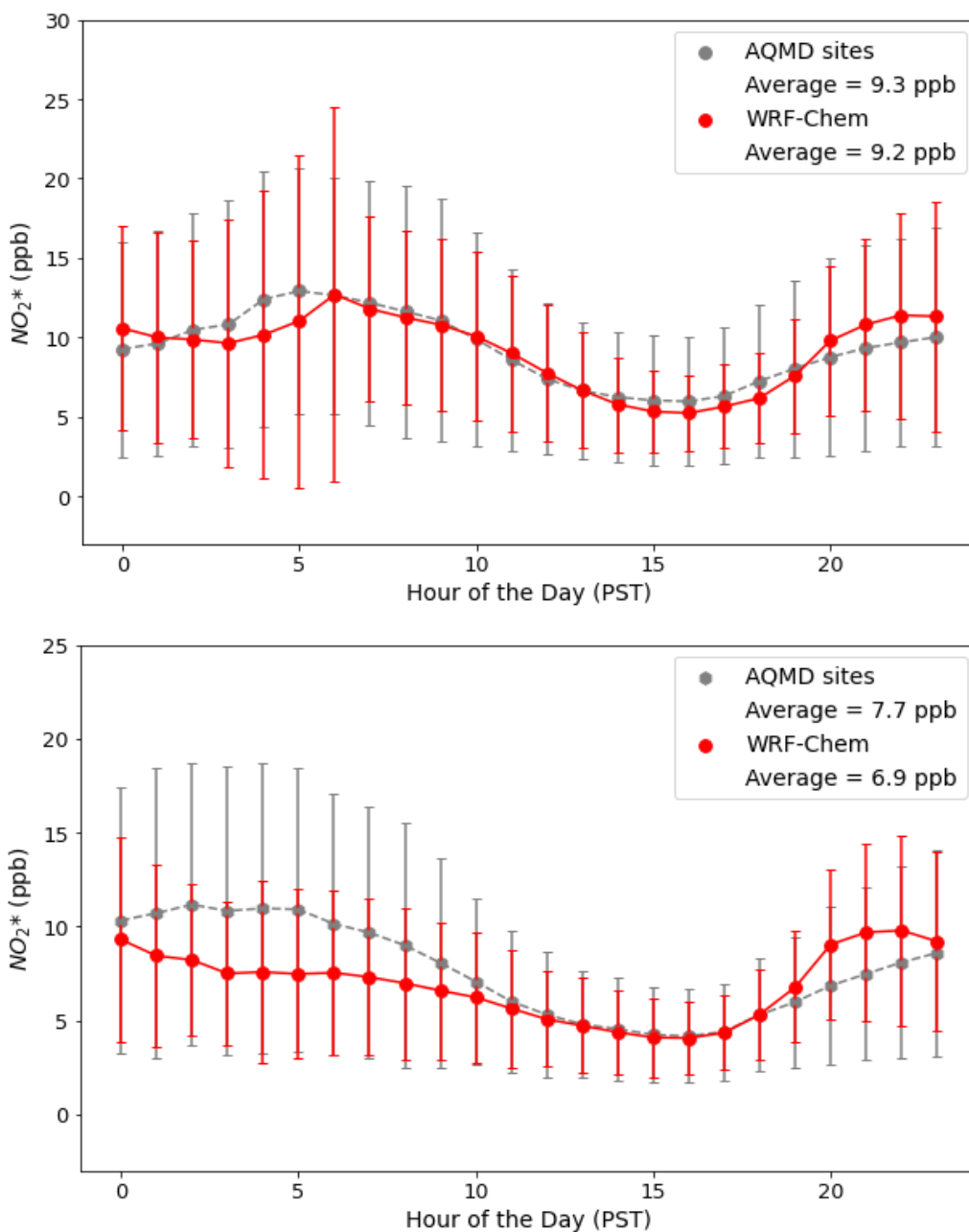


Figure 3.4: Diurnal variation of NO_2^* ($\text{NO}_2^* = \text{NO}_2 + \text{PAN} + \text{alkyl nitrates} + \text{HONO} + 2 \cdot \text{N}_2\text{O}_5$) from WRF-Chem compared to NO_2 from 24 AQMD surface monitors in the South Coast Air Basin on weekdays (top) and weekends (bottom) for June 2021. Error bars show one standard deviation.

Figure 3.5 compares spatial distributions of modeled (WRF-Chem) tropospheric column-integrated NO_2 concentrations with corresponding TROPOMI satellite-derived values. Both model and satellite data show high weekday NO_2 columns over downtown LA extending south towards Long Beach as well as extending further inland to the east as far as San Bernardino. In both cases, there is significantly lower NO_2 on the weekends, especially apparent within the South Coast Air Basin along the corridors east and south of downtown LA. Elevated NO_2 concentrations due to traffic along major highways running through rural areas are captured well, notably on Interstate highways I-5 (heading north towards San Francisco and Sacramento) and I-15 (heading northeast towards Las Vegas) (Figure 3.6). The main difference between modeled and observed NO_2 in the southern California domain is that overall, WRF-Chem has higher NO_2 concentrations in the downtown region, with maximum NO_2 concentrations slightly offset to the northeast compared to what the TROPOMI satellite observations show (Figure 3.7). These differences are slightly emphasized on weekdays in comparison to weekends.

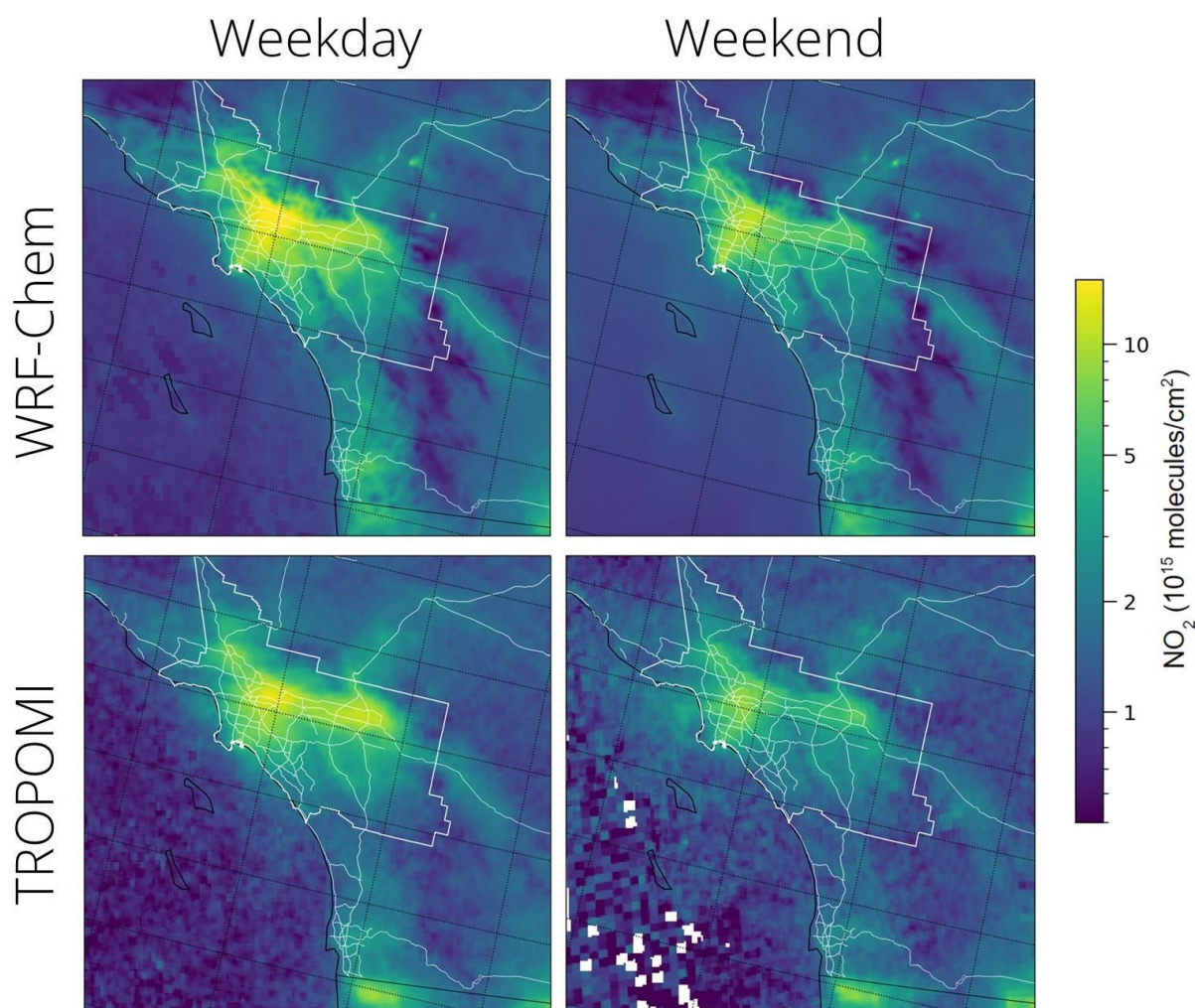


Figure 3.5. Average tropospheric NO_2 columns over southern California for June 2021 predicted using the WRF-Chem model (top row) with comparisons to TROPOMI satellite data (bottom row). Separate results are shown for weekdays (left panels) and weekends (right panels). The boundaries of the South Coast air basin and major highways are shown in white.

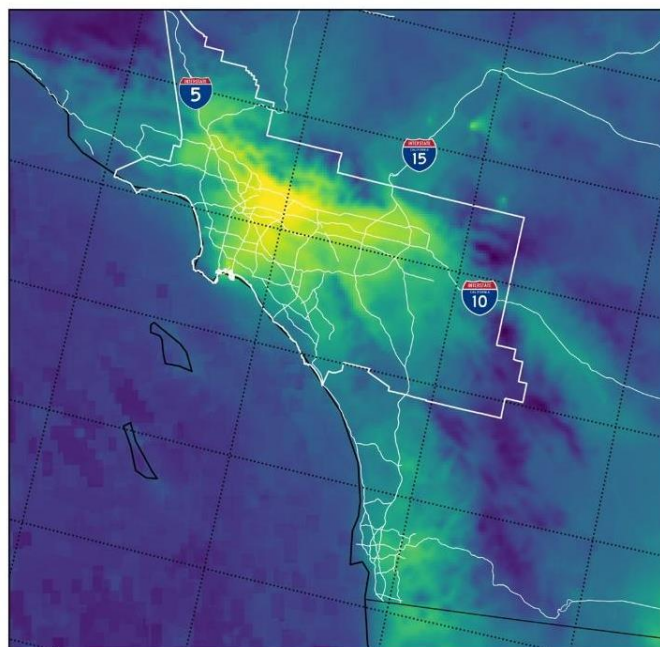


Figure 3.6: Key interstate freeways within the Los Angeles domain for spatial reference

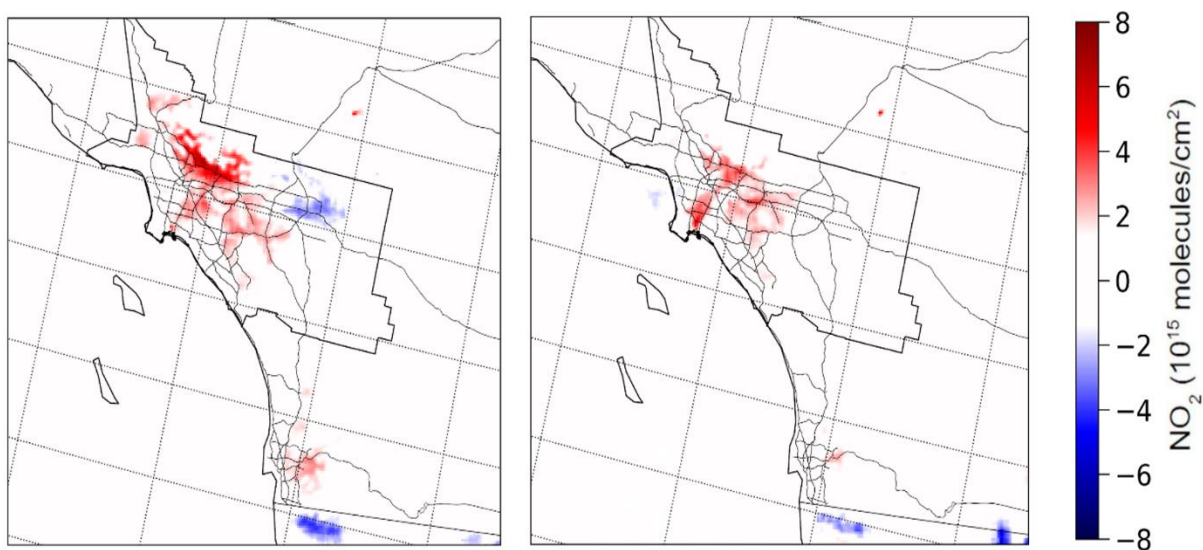


Figure 3.7: Average WRF-Chem vertical NO₂ columns minus average TROPOMI vertical NO₂ columns on weekdays (left) and weekends (right) for June 2021.

Figure 3.8 shows scatterplots of modeled tropospheric NO₂ columns versus corresponding satellite-derived values. The lower NO₂ values that prevail on weekends are apparent in the more limited range of the data in the rightmost panel of Figure 4. The regression-derived coefficients are similar for all three plots, with near-zero intercepts, and slopes of 1.18-1.20. The model explains a high fraction of the observed variance in NO₂ columns, though the value of R² decreases somewhat from 0.89 on weekdays to 0.81 on weekends. The mean normalized bias for the model relative to satellite data is +8% for all days in June, with similar values for the subsets of weekdays only and weekend days only. The finding of slight overestimation in the model compared to the satellite data is in contrast with the finding of slight underestimation compared to aircraft observations. The differences between the model and the satellite are all within the range of and directionally consistent with findings of previous studies that suggest a negative bias of ~20% in satellite-derived observations of NO₂ columns in urban areas.¹⁰⁴

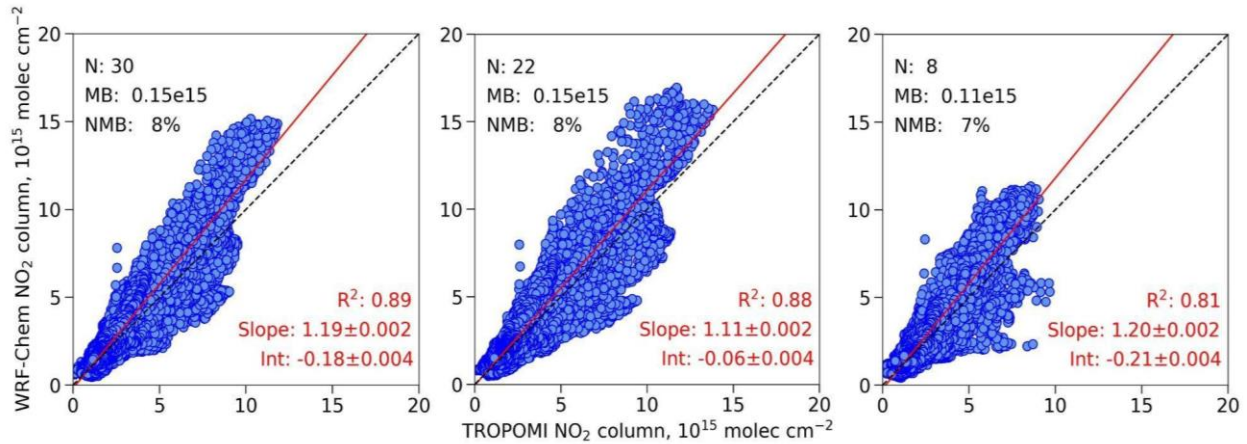


Figure 3.8. Orthogonal distance regression between modeled and satellite-derived tropospheric NO₂ columns over southern California in June 2021 for all days (left panel), weekdays only (middle panel), and weekends only (right panel).

The direct comparison between the model and satellite NO₂ columns at 4 km resolution over the larger California domain can be seen in Figure 3.9. Model performance for our study period at a 4 km resolution for California is comparable to model performance at 1.3 km resolution for southern California within the same domain, as shown in Figure 3.10. At 4 km resolution, we see an R² of 0.83 with a slope of 1.14 for the entire California/Nevada domain and an R² of 0.91 and a slope of 1.09 for the Southern California domain. While the results have similar statistics, the 1.3 km output has the added benefit of resolving sharp gradients in NO₂ over major cities and highways, in line with what can be resolved by the satellite.

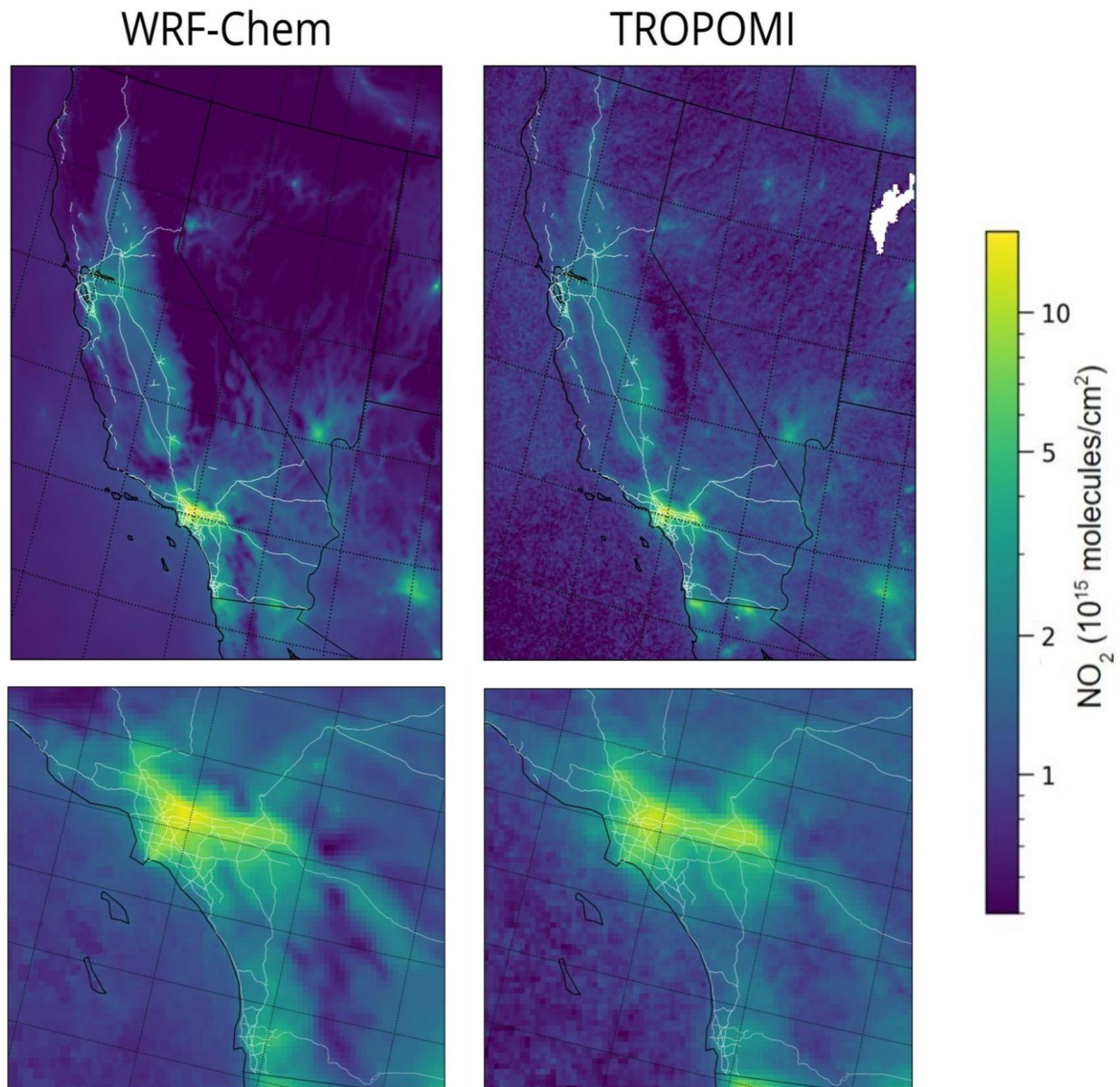


Figure 3.9: Average tropospheric NO₂ columns at 4 km resolution over California for June 2021, predicted using the WRF-Chem model with comparisons to TROPOMI satellite data. The top panels show the entire CA/NV domain, while the bottom panels show the zoomed-in Los Angeles domain. Major highways are shown in white.

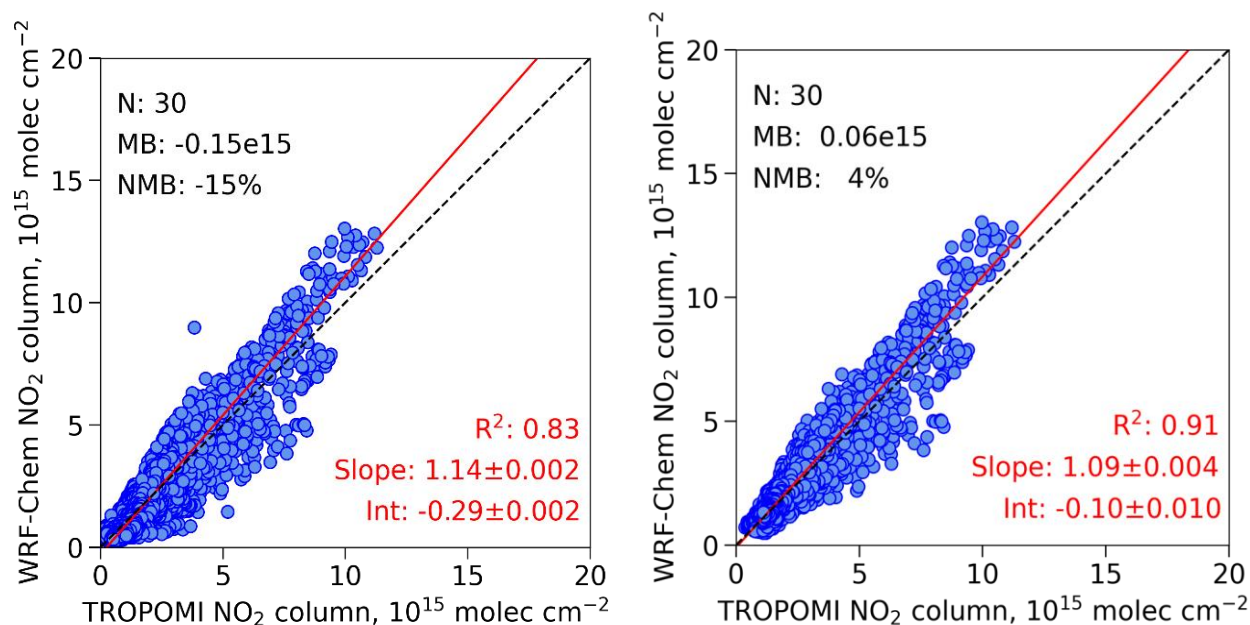


Figure 3.10: Orthogonal distance regression between modeled and satellite-derived NO₂ columns at 4 km resolution in the full California domain (left panel) and the southern California domain (right panel) in June 2021.

To further evaluate the emission inventory, we turn to a more in-depth consideration of weekday-weekend differences in NO₂ columns. Figure 3.11 shows weekday-weekend difference plots of NO₂ columns for the model and the satellite data. The spatial patterns of areas showing large weekend decreases in these plots are similar. The model shows larger weekend NO₂ decreases northwest of downtown along the I-5 Interstate highway through the San Fernando Valley, while the satellite data indicates larger weekend NO₂ decreases further inland, extending to the east as far as San Bernardino. The traffic count data underlying the fuel-based inventory used in this study are for 2018, and may not adequately reflect expansions in warehousing, truck traffic, and associated NO_x emissions that have recently occurred in inland portions of the LA basin.^{119,120}

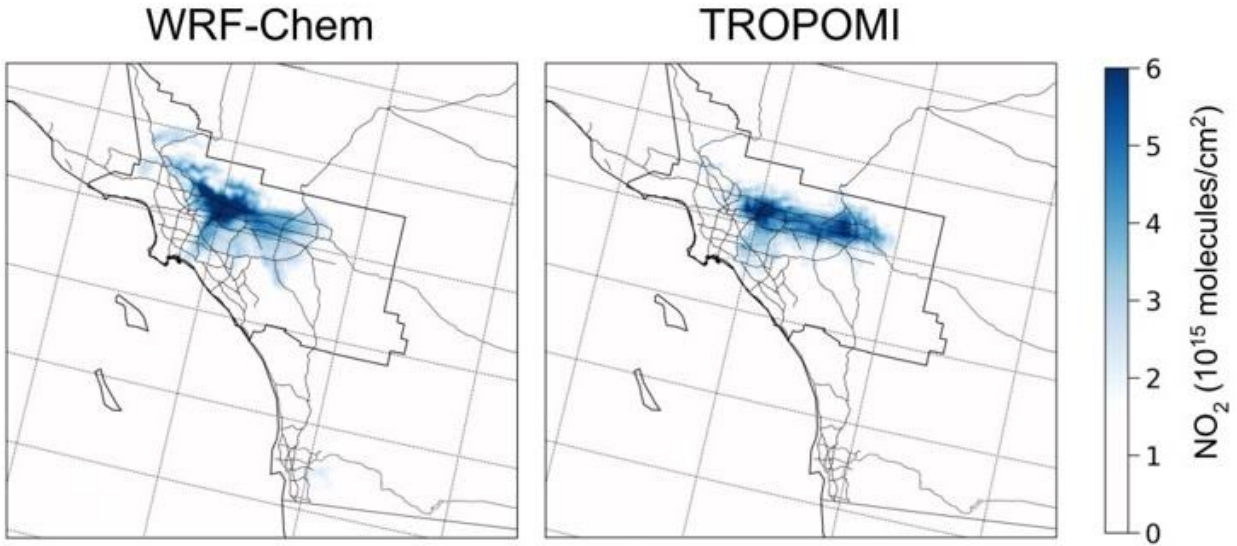


Figure 3.11. Weekday-weekend difference plots for tropospheric NO₂ columns for the WRF-Chem model and TROPOMI satellite data.

An additional emissions sensitivity case was considered in a model run with all on-road vehicle emissions zeroed out completely. Emissions from other source categories were left unchanged. As shown in Figure 3.12, the spatial pattern in the resulting weekday-weekend difference plot for this case without the on-road sector does not match either the observed spatial distribution or the magnitude of weekend NO₂ decreases shown in Figure 3.11. We conclude that locations that exhibit large weekend NO₂ decreases (i.e., from Downtown LA east to San Bernardino) are those where on-road vehicle emissions make large absolute and relative contributions to tropospheric NO₂ columns.

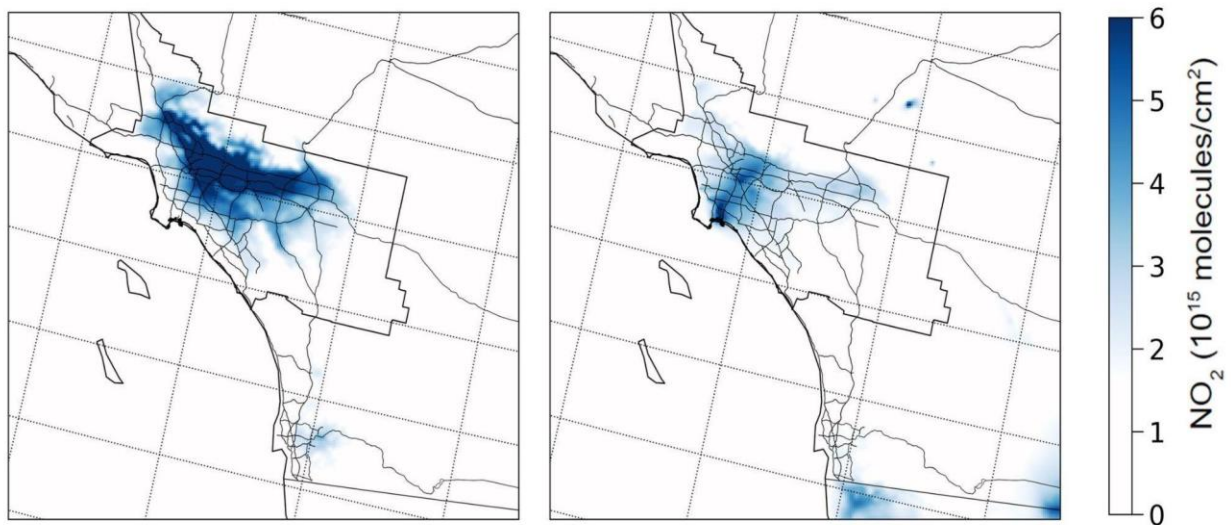


Figure 3.12: Weekday-weekend tropospheric NO₂ column difference of the on-road sector (left) and the remaining combined offroad, area, and point sectors (right)

Current emission inventories used for air quality planning purposes suggest that on-road vehicle emissions are no longer the dominant source of NO_x emissions in southern California. In contrast, as shown in Table 3.3, the emission inventory in the present study has 25% higher NO_x emissions overall when compared to the planning inventory, with the dominant contribution coming from on-road sources. More specifically, the present study includes higher emissions from on-road vehicles (198 ± 69 versus 103 tons/day) that account for 66% of anthropogenic NO_x emissions. The planning inventory assigns greater importance to area and off-road mobile sources, which together account for a higher fraction (49 versus 26%) of total NO_x emissions compared to the present study.

Table 3.3. Comparison of NO_x Emission Inventories for the South Coast Air Basin

	Fuel-based Inventory June 2021		South Coast Air Quality Management District, Summer 2022*	
Source Sector	NO _x (tons/day)	fraction of total	NO _x (tons/day)	fraction of total
on-road	198 ± 69	66%	103	43%
area + off-road	79	26%	116	49%
point	23	8%	20	8%
Total	300		239	

*Ocean-going vessels removed to match included sectors of fuel-based inventory.

Air quality model results, satellite observations, and their corresponding weekday-weekend differences are consistent with a larger-than-expected on-road vehicular source of NO_x emissions in southern California. Major efforts to update fleets to newer engine models with advanced emission control systems have improved heavy-duty diesel, and values from the EMFAC model are consistent and within the uncertainty of those from the fuel-based inventory evaluated here. Still, actual emissions from heavy-duty diesel may be even higher than estimated due to ineffective NO_x control during idling and low-load driving conditions. Total NO_x emission estimates are higher in this study for on-road vehicles in comparison to the EMFAC inventory, owing to higher emissions from gasoline vehicles and light- and medium-duty diesel vehicles. Contributing factors to these differences may include slow absolute progress in reducing fleet-average NO_x emission factors for gasoline vehicles since 2010 (see Figure 2.5 in Chapter 2) and slow fleet turnover with relatively high in-use NO_x emission factors for light- and medium-duty diesel trucks.

Chapter 4: Projecting Spatial Distributions of Pollutants and Health Effects

4.1 Introduction

Fine particulate matter (PM_{2.5}) has been a focal point of air quality reduction strategies globally, primarily due to its notable risk to human health. The small particle size allows PM_{2.5} to penetrate human defenses and deposit in the lungs, leading to increased mortality and morbidity.^{121,122} While the highest number of PM_{2.5}-related deaths occur in Asia at elevated concentrations, research shows that even lower concentrations of PM_{2.5} observed across North America are associated with adverse health impacts.^{89,123,124} The consequences of widespread PM_{2.5} exposure encompass a broad range of health concerns. Some of the negative health effects that may occur include increased risk of mortality due to cardiopulmonary disease^{8,9} and lung cancer¹⁰. Individuals with pre-existing chronic conditions like asthma are particularly vulnerable to the effects of PM_{2.5}, with increased symptoms and exacerbations of respiratory illnesses for those who experience higher PM_{2.5} exposure.^{11–13}

PM_{2.5} formed from anthropogenic emissions not only poses a significant health hazard to all people but is disparately harmful to communities of color.^{125–129} Recent studies have shown that race-ethnicity exposure disparities are large, with average PM_{2.5} exposure 14% higher for people of color than the average, and with Black people experiencing the largest disparity at 20% higher than the overall population.¹³⁰ Studying how secondary PM_{2.5} exposure changes under projected decreases in NO_x reveals how these disparities may shift over time as well. Recent changes to the national ambient air quality standard in the United States to protect human health are consequential, with the limit for the annual average concentration dropping from 12 to 9 µg/m³ as of February 2024.¹³¹ To reach these limits and reduce the harm caused by PM_{2.5}, it is imperative to address major sources from an emissions standpoint, especially in heavily populated urban centers.

Understanding and addressing emissions of PM_{2.5} and its precursors is challenging due to uncertainties in aggregate emissions at state and national scales as well as significant spatio-temporal variability. PM_{2.5} comes in a wide variety of compositions, is emitted by both natural and anthropogenic sources, and can be sourced through primary and secondary pathways. A key form of secondary PM_{2.5} is ammonium nitrate (NH₄NO₃), which is formed from NO_x and NH₃ reaction pathways. To give perspective on how much PM_{2.5} is formed from NO_x, the global NO_x contribution to PM_{2.5} is estimated to be increasing from 17% in 1990 to 28% in 2012.¹³² In the United States, the fraction of PM_{2.5} attributed to nitrogenous precursors varies greatly based on region and time of year and is estimated to range from 4 to 37% of total PM_{2.5}.¹³³

Within the national context of vehicular contributions to PM_{2.5}, California stands out as a critical region. Recent data highlights the challenge of meeting the new national ambient air quality standard: 29 counties in California exceed the new standard based on 2020-2021 ambient air monitoring data, and 23 of these counties are projected to continue exceeding the 9 µg/m³ threshold in 2032.¹³⁴ Ammonium nitrate is a key contributor to PM_{2.5} levels in California. The contribution of ammonium nitrate to total PM_{2.5} in the San Joaquin Valley can be as high as 30-

80% in the winter season.^{135–137} Given that NO_x is a key precursor to $\text{PM}_{2.5}$, a further examination of how changes to NO_x emissions will affect secondary $\text{PM}_{2.5}$ distributions is critical for understanding the benefits of NO_x reductions and where future emission reduction efforts should be targeted.

The transportation sector is the largest contributor to NO_x emissions in California. This sector is undergoing rapid evolution, with substantial changes anticipated as increasing numbers of vehicles are equipped with advanced emission control technologies. Governor Gavin Newsom's recent Executive Order emphasizes California's commitment to vehicle electrification, setting ambitious goals including 100% of new vehicle sales must have zero emissions by 2035, a full transition to zero-emission short-haul/drayage trucks by 2035, and a full transition to zero-emission buses and long-haul heavy-duty trucks by 2045 where possible.¹³⁸ To understand the role these policy changes and technological advancements will play in reducing air pollution, high-resolution mapping of on-road emissions is needed. Such inventories provide a critical basis for assessing how upcoming technological transitions in the on-road vehicle fleet will contribute to improving human health and reducing exposure to air pollution for different regions.

In the preceding chapter, a high-fidelity Chemical Transport Model (CTM) was employed to gain insights into a specific timeframe during the summer season in 2021.⁹¹ However, the high computational cost associated with coupled weather-chemistry models creates a limitation in applying these tools to model annual average concentrations and to consider larger domains at high spatial resolution. The Intervention Model for Air Pollution (InMAP) offers an alternative. InMAP is a reduced complexity model utilizing a variable-size computational grid that allows for finer spatial resolution in high-population regions.⁵¹ InMAP uses pre-calibrated relationships between changes in emissions and annual average $\text{PM}_{2.5}$ and other pollutant concentrations. This modeling approach allows for the assessment of the impacts of NO_x emissions on secondary $\text{PM}_{2.5}$ over annual time scales. This contrasts with prior results presented in Chapter 3 which addressed summertime conditions for June 2021 and used the high-resolution inventory of NO_x emissions to predict gaseous NO_2 concentrations in comparison with related satellite observations.

Building on the high-resolution on-road NO_x emission inventory developed in the previous chapter, this chapter now explores the influence of trends in emission factors and fleet changes on the spatial distributions of secondary $\text{PM}_{2.5}$ across California. Unique features of the emission inventory, such as high spatial resolution (1.3 km) and inclusion of a light- and medium-duty diesel vehicle category with high NO_x emission factors, are integrated into this analysis. The inventory is expanded to include the entire state and estimates current (2021) as well as future (2030, 2040) NO_x emissions. Assessing resulting NO_x and secondary $\text{PM}_{2.5}$ concentration reductions at up to ~1 km resolution within California's population centers facilitates the identification of key locations to prioritize in vehicle emission reduction efforts. This research not only examines how overall secondary $\text{PM}_{2.5}$ exposure changes under future conditions but also how the emission changes will affect various regions of California and different demographic groups.

4.2 Methods

4.2.1 Spatial and Temporal Expansion of the High-resolution Emission Inventory

The spatial extent of the existing high-resolution motor vehicle emission inventory is extended beyond the regional focus on the Los Angeles area in Chapter 3 to encompass all of California. The emission mapping methodology used here follows the methods described in Chapter 3 but now uses a statewide 1.3 km x 1.3 km grid.¹³⁹ The inventory assigns the majority of taxable sales (i.e., fuel intended for on-road use) of gasoline (68%) and diesel fuel (77%) in California⁵⁹ using 2018 FHWA traffic count data for highways and arterial roadways. The remaining fuel use is apportioned to grid cells based on census block-level population estimates.^{32,77} Previous fuel-based inventories separated vehicles into only two categories: light-duty gasoline vehicles and heavy-duty diesel vehicles. A key feature of this inventory is the assignment of 22% of on-road diesel fuel use to a third category, consisting of light- and medium-duty diesel vehicles. These vehicles were determined previously (see Chapter 2) to have higher NO_x emission factors than heavy-duty diesel trucks, and a more slowly decreasing trend over time.⁹¹ Future fuel consumption for each vehicle category is projected using EMFAC-derived ratios of fuel consumption in the future (2030 and 2040) versus baseline (2021) years at the county level, accounting for counties that are split between more than one air basin.⁷⁶

Total statewide taxable fuel sales are distributed to road segments based on vehicle counts and are combined with fuel-based emission factors (mass of NO_x emitted per unit of fuel burned). The emission factors are derived from regression analyses of roadside emission spectrometer and highway tunnel measurements (see Chapter 2). To predict unmeasured future year emission factors, a modified version of the regression-based analyses from Chapter 2 was developed for gasoline and light-/medium-duty diesel.⁹¹ These fits were modified to be logarithmic, transitioning from polynomial and linear fits to measured data. This modified fitting procedure accounts for measured values that continue to decrease but at slower absolute rates as the vehicle fleet becomes cleaner on average. Linear and polynomial fits to measured emission factor trends gave unrealistic (e.g., negative or increasing) results for emission factors when extrapolating from historical and present-day values 10-20 years into the future. The modified emission factor fitting procedure does not significantly affect our estimates of baseline emissions in 2021, with a difference of <5% in NO_x emissions overall. Higher than usual emissions of NO_x and other pollutants, such as CO and VOC, are known to occur at the beginning of vehicle trips when the engine and catalytic converter are not yet warmed up. These excess emissions are referred to as cold-start emissions and are estimated to account for 25% of total NO_x emissions from gasoline vehicles. Cold start emissions are accordingly incorporated into the emission factors values for gasoline engines, consistent with previous studies.

For heavy-duty diesel truck engines, emission factors are scaled to reflect conditions in 2030 and 2040 using EMFAC-derived emission factor forecasts at the county level instead of the linear trendline used previously in Chapter 2.⁷⁶ An effective NO_x emission factor from EMFAC is calculated for each year by dividing estimated NO_x emissions by total fuel consumption for each county. Forecast ratios between NO_x emission factors in the future (2030 and 2040) and the baseline (2021) year are used to scale the baseline heavy-duty diesel emission factor from Chapter 2 to derive future-year emission scenarios. This approach acknowledges ongoing

significant changes to heavy-duty vehicle emission control and anticipates that the linear trend used to represent past improvements may not be representative moving forward in time. More rapid fleet turnover to SCR-equipped trucks and deterioration of current SCR systems are some of the factors that could lead future-year emission factors to quickly deviate from the current rapid downward trajectory. Decreases in NO_x emissions due to vehicle electrification are implicitly accounted for in the underlying fuel consumption amounts, which show slower growth as more and more vehicles transition to electric powertrains. The resulting NO_x emission factors used to construct the high-resolution on-road vehicle emission inventory are shown in Figure 4.1.

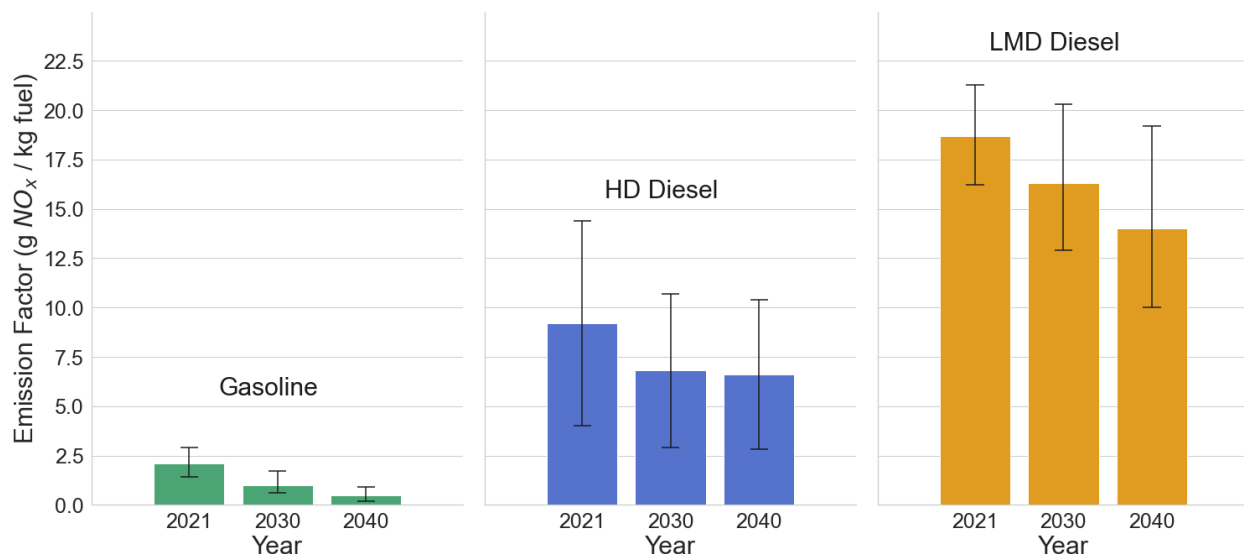


Figure 4.1: Emission factors trends (g NO_x emitted/kg fuel burned) for each of three vehicle categories in California: gasoline engines, heavy-duty diesel trucks, and light- and medium-duty diesel vehicles. Error bars indicate a 95% confidence interval.

4.2.3 Intervention Model for Air Pollution (InMAP)

Because steep computational cost remains a significant barrier to using chemical transport models for high-resolution modeling of annual averages and larger spatial regions, this study utilizes the Intervention Model for Air Pollution (InMAP) to project annual average changes to NO_x and secondary PM_{2.5} concentrations for California.⁵¹ InMAP is a reduced-complexity model that predicts how changes to primary pollutant emissions affect spatial patterns of air pollutant concentrations and associated human exposures. With a variable grid with the highest resolution (1 km x 1 km grid cells) in heavily populated regions collocated with high road density, the model is well-suited to consider the effects of changes in vehicle emissions which are concentrated in urbanized areas. Conversely, the lowest resolution grid cells (48 km x 48 km) are used over large areas of the state that are sparsely populated. While the present analysis only considers the effects of emission changes that occur within California, the air quality modeling of impacts extends to include all of the continental US, including impacts on neighboring states from pollutant emissions that occur within California.

To mitigate significant computational costs, InMAP deviates from traditional CTMs in several ways. Instead of simulating physical and chemical variables at each time step based on first principles and empirical representations of atmospheric processes, InMAP solves for the steady-state solution to the standard reaction-advection-diffusion equation using parameterizations for chemistry and physics derived from the output of a year-long CTM run. The foundational physical and chemical information in this version of InMAP is derived from a 12 km resolution CTM run of the continental US for the year 2006, as described in Tessum et al. 2015.¹⁴⁰ At each time step, the algorithm calculates the change in concentration based on the physical and chemical parameterizations for each grid cell derived from this WRF-Chem run. Advection is parameterized using Reynolds decomposition consisting of an annual average, deviations from the annual average resolved in WRF-Chem, and deviations from the annual average that are not resolved in WRF-Chem. An annual average partitioning coefficient for each grid cell quantifies the conversion of NO_x emissions to particulate nitrate. Modeling secondary particulate nitrate on an annual average basis is particularly challenging due to seasonal patterns of higher particulate nitrate formation under cold, stagnant winter conditions.¹⁴¹ While InMAP does not estimate particulate nitrate as well as primary PM_{2.5} in comparison to the WRF-Chem model, model-measurement comparisons at a national scale have shown InMAP particulate nitrate estimates still meet established air quality model PM_{2.5} performance criteria (mean fractional bias $\leq \pm 60\%$ and mean fractional error $\leq 75\%$).⁵¹ More details and a comprehensive overview of the model parameterizations and performance can be found in Tessum et al. 2017.⁵¹

The input data used for the InMAP model runs is the 1.3 km resolution gridded emission inventory for all of California described in the previous section. Because InMAP is designed to use marginal changes in emissions, the inputs are formatted as changes in NO_x emissions between the baseline year (2021) and each of two future years (2030 and 2040). While InMAP processes emissions of NO_x, VOCs, SO_x, NH₃, and primary PM_{2.5}, all emission changes in this study are set to zero except for NO_x emitted by vehicles. Consequently, the resulting changes to secondary PM_{2.5} concentrations are exclusively changes to particulate nitrate due to NO_x emission decreases in the vehicle sector. NO_x emissions are input in the ground-level model layer in InMAP, which is appropriate for on-road vehicle sources. The demographic data used to determine population exposure by race is sourced from the 2010 Census Data at the block level, aggregated into the following categories: non-Hispanic White, Latino, Black, Asian, and Native. In California, these groups account for 35, 40, 6.5, 16, and 1.7% of the total population respectively¹⁴²

4.2.4 Health Impact Calculations

Health impacts for this study are assessed based on changes in premature mortality (death) associated with changes in secondary PM_{2.5} exposure resulting from the decreases in NO_x emissions from on-road motor vehicles. Exposure is quantified as the population-weighted annual average concentrations of secondary PM_{2.5} calculated using Eq. 4.1:

$$C_i = \frac{c_i \cdot p_i}{\sum p_i} \quad \text{Eq. 4.1}$$

In this equation, C_i is the population-weighted annual average concentration in $\mu\text{g}/\text{m}^3$ for the target demographic within the grid cell i , c_i is the annual average concentration within the grid cell, and p_i is the population of the target demographic. By multiplying the concentration change in the grid cell by the cell population and dividing by the total population, higher exposure is attributed to more populous grid cells.

Excess deaths avoided by the decrease in secondary PM_{2.5} are calculated using a Cox proportional-hazards model (Eq. 4.2), as described in Wang et al. 2023 and Tessum et al. 2019.^{130,143}

$$D_i = m_i \cdot p_i \cdot e^{\frac{\log(h)}{10} \cdot c_i - 1} \quad \text{Eq. 4.2}$$

In this equation, D_i is the annual excess deaths in grid cell i , m_i is the baseline mortality rate, p_i is the population of the target demographic within the grid cell, c_i is the concentration of secondary PM_{2.5}, and h is the hazard ratio. The hazard ratio signifies the increased mortality from each respective cause associated with exposure to an increase of 10 $\mu\text{g}/\text{m}^3$ in long-term PM_{2.5} exposure. Appropriate values for the hazard ratio are determined from Krewski et al. 2009. For this study, the hazard ratios used are 1.078 for deaths from all causes, 1.128 for cardiopulmonary disease (CPD), 1.287 for ischemic heart disease (IHD), and 1.142 for lung cancer.¹⁴⁴ The base PM_{2.5} concentration from InMAP is added to the marginal changes to account for the slight non-linearity of this calculation, where health impacts are higher at higher concentrations. The uncertainty from using the older baseline PM_{2.5} concentrations is minor, as the non-linearity is slight at the relatively low concentrations seen in North America. For calculations of marginal deaths accompanying the decrease in emissions, the premature deaths associated with the projected 2040 secondary PM_{2.5} contribution of vehicles are subtracted from the premature deaths associated with the 2021 secondary PM_{2.5} contribution of vehicles.

4.3 Results and Discussion

The gridded NO_x emission inventory for on-road vehicles in California is presented in Figure 4.2. This figure provides a comprehensive overview of the spatial distribution of on-road vehicle activity and emissions. Inset maps for the San Francisco Bay Area/Central Valley and Southern California are presented in separate panels. Emissions are highest in major metropolitan areas, particularly the Bay Area and Los Angeles, due to the high associated densities of population and major roadways and highways. Other areas with high densities and levels of NO_x emissions include Sacramento, San Diego, and along Interstate 5 running north-south through the state. To quantify the magnitude of the emissions, Table 4.1 provides a summary of total yearly NO_x emissions from on-road vehicles in this study in the baseline year (2021) as well as projections for two future years (2030 and 2040). The South Coast Air Basin has the highest overall NO_x emissions in all years, well above emissions levels in the San Francisco Bay area which ranks second in all years. When compared to baseline emissions in 2021, emissions of NO_x from on-road vehicles decrease by 38% in 2030 and by 55% in 2040. The relative decreases in emissions for individual air basins vary between 30-45% in 2030 and between 35-66% in 2040. These intrastate differences reflect regional variations in expected growth in population and vehicle travel, as well as expected adoption rates of electric vehicles. More rural areas such as the San Joaquin Valley have slower electric vehicle adoption rates when compared to statewide averages and California zip codes with higher income, percentages of White and Asian people, and house costs have higher rates of electric car ownership.^{145,146} The affordability of new electric vehicles and accessibility of charging infrastructure remain key obstacles to more widespread adoption across all communities.

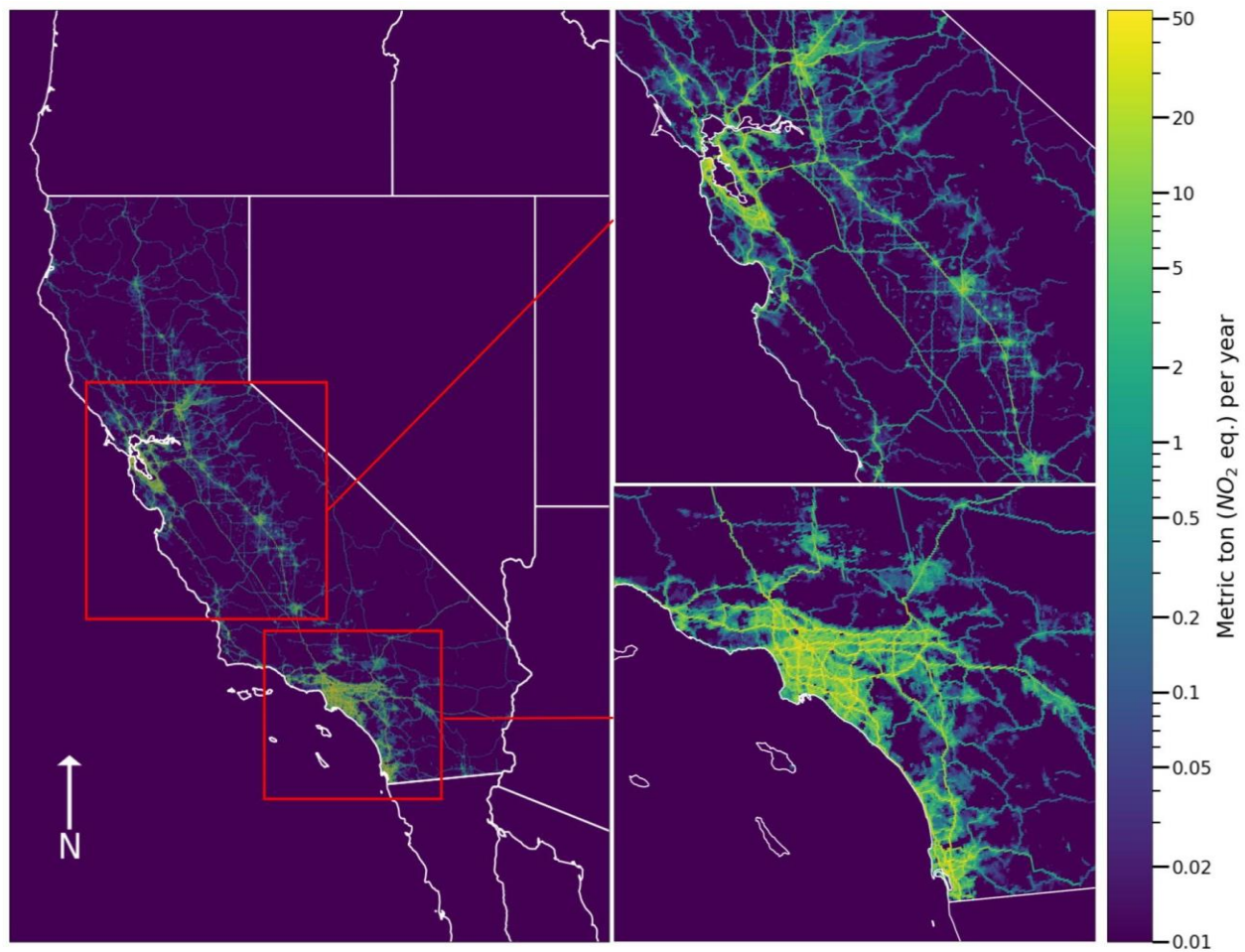


Figure 4.2: Spatially resolved decrease in annual on-road vehicle emissions of NO_x between one of the future years (2040) and baseline conditions (2021), in metric tons per year. Inset panels on the right show results for the San Francisco Bay Area/Central Valley and Los Angeles area in more detail.

Table 4.1: Annual average NO_x emission rates from on-road vehicles in each California Air Basin in units of metric tons (NO₂ eq.) per day.

Air Basin	2021	2030	2040
South Coast	222	138	96
San Francisco Bay	104	63	42
San Joaquin Valley	73	46	38
San Diego County	53	32	23
Sacramento Valley	48	29	21
Mojave Desert	29	19	18
South Central Coast	26	14	9.4
Salton Sea	13	9.1	8.5
North Central Coast	13	7.5	5.0
Mountain Counties	12	6.9	5.1
North Coast	7.5	4.2	2.9
Northeast Plateau	3.9	2.3	1.8
Great Basin Valleys	2.3	1.4	1.1
Lake County	1.3	0.7	0.4
Lake Tahoe	0.9	0.5	0.3
Statewide Total	608	375	272

Figure 4.3 presents a comparison of statewide estimates of NO_x emissions from the fuel-based inventory and the EMFAC model. There is an evident discrepancy between the two inventories for the baseline year (EMFAC is much lower), consistent with the substantial underestimate of NO_x emissions identified in the previous chapter for the South Coast Air Basin. Statewide for baseline (2021) conditions, the fuel-based inventory estimates for NO_x are 63% higher than corresponding estimates from the EMFAC model. In future years 2030 and 2040, the relative difference in estimates narrows to 43 and 35%, respectively, indicating gradual convergence in the emission estimates over time. When examining emissions by vehicle type, an interesting pattern emerges. Emissions from gasoline and heavy-duty diesel vehicles converge to similar values overall and demonstrate comparable rates of decrease over time. As the vehicle fleet rapidly electrifies and high emitters are phased out, differences in emission factors are reduced and the effect of these differences has less impact on total emissions. By 2040, estimates of NO_x emissions from gasoline engines especially align closely in both inventories.

A noticeable difference in NO_x emission estimates for the light- and medium-duty diesel vehicle category persists for all years shown in Figure 4.3. In the EMFAC inventory, this vehicle category experiences an extremely rapid NO_x emission decrease compared to the fuel-based inventory. This substantial gap in emission estimates raises questions about the assumptions regarding the rate of fleet turnover and the effectiveness of emission control technologies for diesel vehicles in the EMFAC inventory. The measured trend in NO_x emission factors from the light- and medium-duty diesel vehicle category indicates a slow rate of progress over the last decade; such a steep rate of decrease is not supported by the observed past rate of improvement in on-road measurements. Policy changes that require the light- and medium-duty diesel vehicle sector to upgrade to SCR-equipped engines or transition to electric powertrains could accelerate the rate of NO_x emission reduction beyond what the historical record has shown in this sector to date.

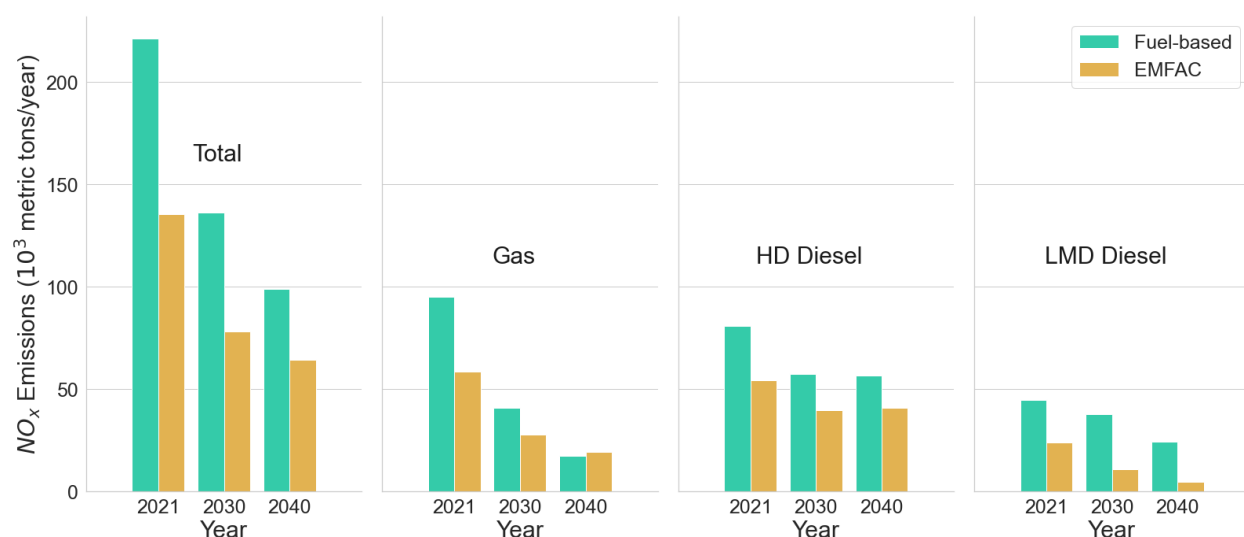


Figure 4.3: Comparison of trends in total statewide NO_x emissions by on-road vehicle category for the fuel-based inventory and California's EMFAC model for baseline and future years.

Figure 4.4 shows statewide maps of anticipated reductions in secondary PM_{2.5} and NO_x concentrations stemming from projected reductions in NO_x emissions between 2040 and 2021. Differences between 2030 and 2021 are not shown in Figure 4.4, but they exhibit a similar spatial pattern with smaller absolute magnitudes for air quality improvements since the emission reductions are not as large. Overall, the decreases in NO_x concentrations across the state are localized and primarily observed in close proximity to populated areas where the majority of NO_x is being emitted. In contrast, decreases in secondary PM_{2.5} concentrations resulting from emissions changes are more widespread, extending into neighboring states, such as Nevada and Arizona. Decreases in air pollution are seen in all areas with major freeways for both pollutants. However, the regions experiencing the largest decreases differ depending on the pollutant. For NO_x, there are significant reductions along all major freeways, with the largest decreases in major urban centers such as Los Angeles and the San Francisco Bay Area where road density is highest. Conversely, for secondary PM_{2.5}, larger air quality improvements are seen within the Central Valley along Interstate 5 and Highway 99 corridors, and in San Diego, rather than in the more highly populated urban areas mentioned above.

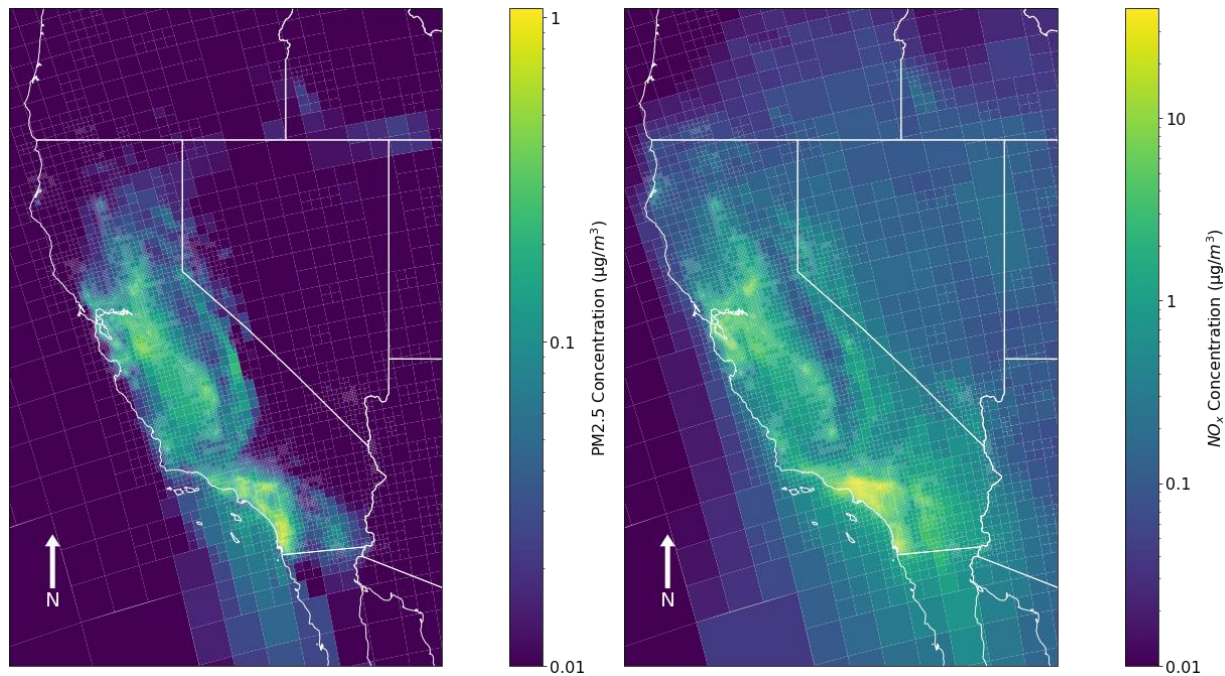


Figure 4.4: Statewide decreases in annual average secondary PM_{2.5} concentrations (left) and NO_x concentrations (right) in 2040, calculated relative to 2021 baseline values.

Figure 4.5 zooms in on regional-scale changes in secondary $\text{PM}_{2.5}$ and NO_x concentrations between 2021 and 2040 for Southern California and the San Francisco Bay Area and Central Valley. Predicted secondary $\text{PM}_{2.5}$ concentration changes in Southern California exhibit an unexpected spatial pattern, with larger decreases observed further south towards San Diego, whereas the largest NO_x decreases occur in the Los Angeles area. A similar though less pronounced effect occurs in the Bay Area, where the decrease in secondary $\text{PM}_{2.5}$ is smaller within the densely populated urban core, and larger at comparatively less densely populated locations further inland. These larger-than-expected decreases in secondary $\text{PM}_{2.5}$ in downwind/less populated areas may indicate ammonium nitrate formation is more strongly NO_x limited in San Diego and the Central Valley. For the Central Valley, this finding is consistent with high emissions of the other relevant precursor (ammonia) from agricultural sources such as fertilizer use and livestock. Other contributing factors may include faster NO_x to NH_4NO_3 conversion rates in those regions and significant transport of NO_x emissions inland to the Central Valley and southward towards San Diego.

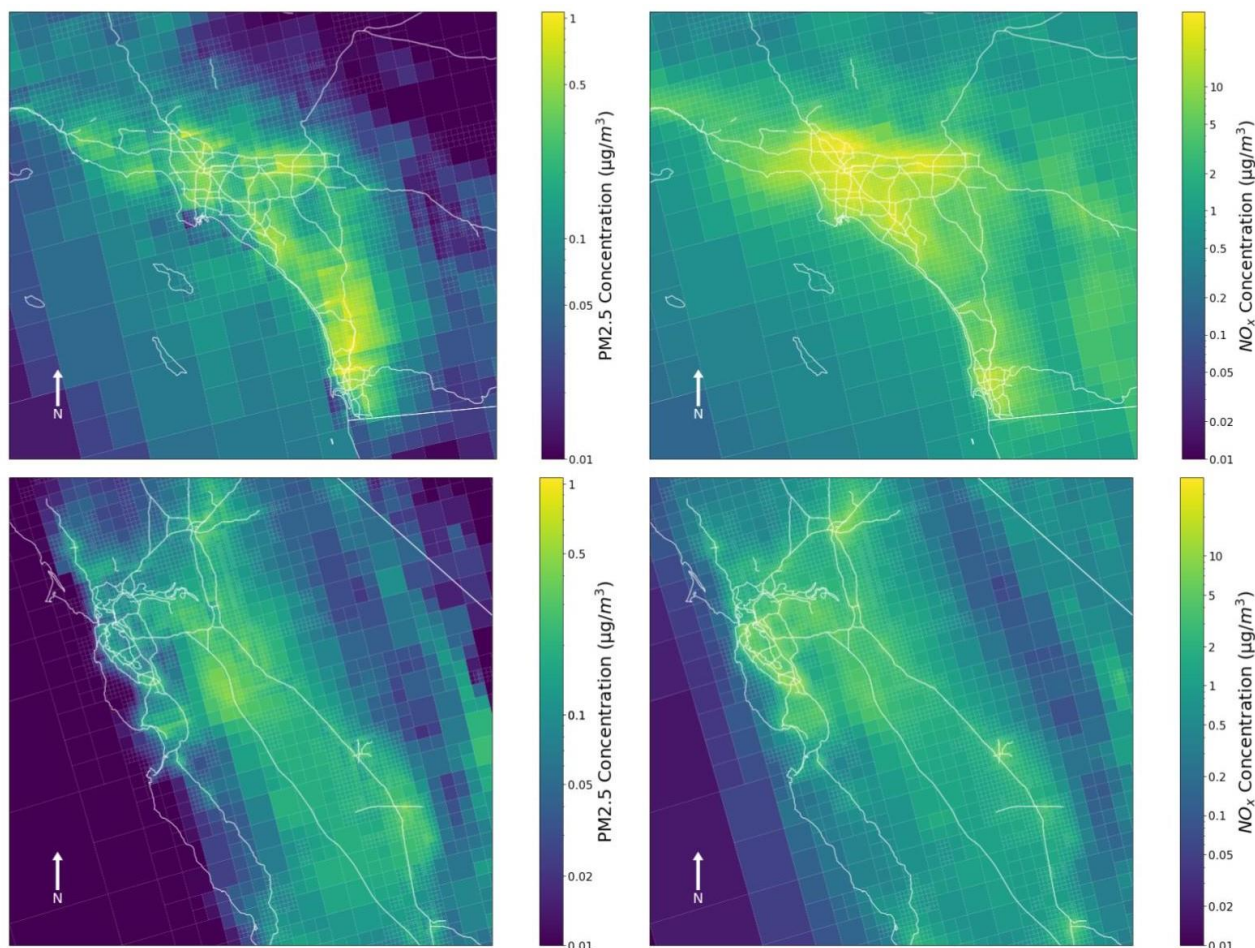


Figure 4.5: Projected decreases in annual average secondary $\text{PM}_{2.5}$ (left) and NO_x concentrations (right) in 2040 compared to baseline 2021 values. The upper panels show results for Southern California, while the lower panels focus on the Bay Area and the San Joaquin Valley.

Figure 4.6 shows reductions in population-weighted exposure at the air basin scale, with the total population denoted as a black line and results plotted separately for each racial demographic group. A universal decline in secondary PM_{2.5} exposure is observed for all air basins, and a larger decrease is seen in higher-population air basins such as San Francisco Bay, South Central Coast, Sacramento Valley, San Joaquin Valley, South Coast, and San Diego County. San Diego County has the largest population-weighted exposure decrease of 0.43 $\mu\text{g}/\text{m}^3$. The two most populous air basins (i.e., South Coast and San Francisco Bay area) also see decreases in population-weighted exposure of particulate nitrate of 0.29 and 0.14 $\mu\text{g}/\text{m}^3$, respectively.

The San Joaquin Valley, a pivotal area of interest in NO_x reduction initiatives, shows the third largest decrease in population-weighted exposure at 0.22 $\mu\text{g}/\text{m}^3$. Small variations are seen within air basins, with Latino, Asian, and Black populations projected to see slightly larger decreases in exposure relative to the total and White and Native populations projected to see slightly smaller decreases relative to the total. However, the differences shown in the current study are minor overall. While the modeled results may not exhibit clear exposure differences at this scale it is important to recognize the potential for patterns to be obscured by simplified modeling techniques and spatial aggregation, particularly for historically segregated and redlined communities. As model performance becomes more reliable at high resolution, identifying the effects on those communities is of utmost importance. The results shown here not only underscore the positive trajectory in air quality improvements across air basins, but also highlight the potential need for targeted emission reduction strategies that consider demographic variations to ensure equitable progress in reducing population exposure to secondary PM_{2.5}.

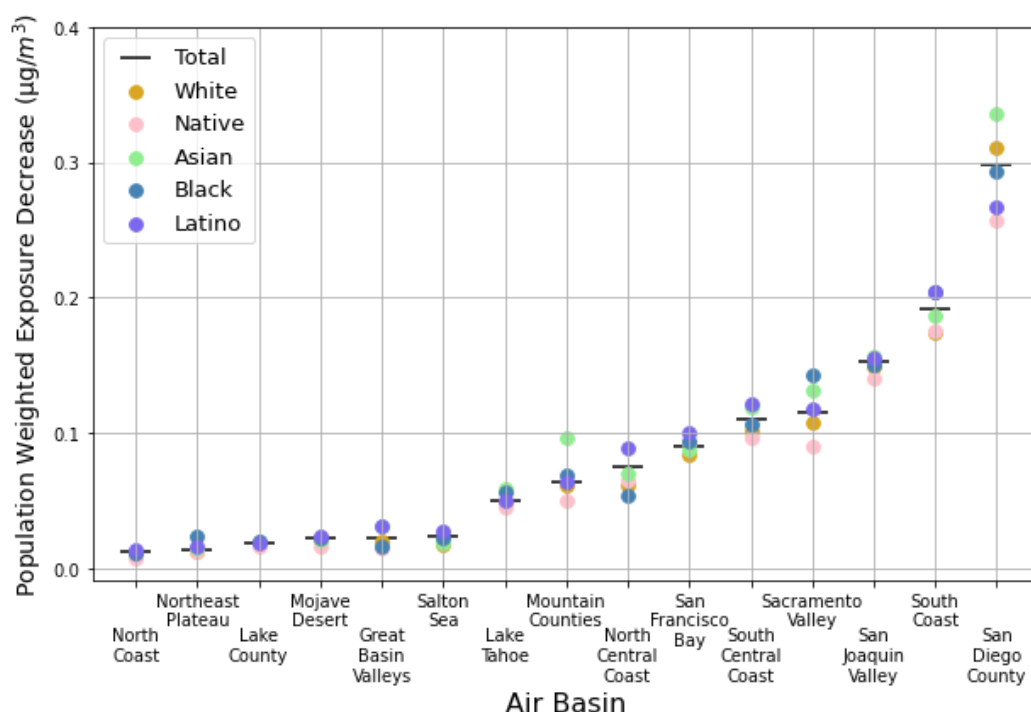


Figure 4.6: Projected population-weighted decreases in secondary PM_{2.5} exposure between 2021 and 2040 by air basin, for the total population and White, Native, Asian, Black, and Latino subpopulations in each air basin.

Figure 4.7 shows regionally averaged decreases in secondary $\text{PM}_{2.5}$ exposure (left panel), which roughly mirrors the spatial distribution of avoided premature deaths from all causes associated with secondary $\text{PM}_{2.5}$ exposure changes (right panel). While there is general alignment between these metrics, differences arise because more premature deaths occur in areas with higher populations, regardless of whether these areas see the largest decreases in population-weighted exposure. The South Coast Air Basin experiences the highest reduction in expected premature deaths despite the finding that San Diego will see a larger reduction in population-weighted exposure to secondary $\text{PM}_{2.5}$ overall. Similarly, the San Francisco Bay Area will also see a larger reduction in premature deaths despite lower relative reductions in population-weighted exposure to air pollution than the San Joaquin Valley.

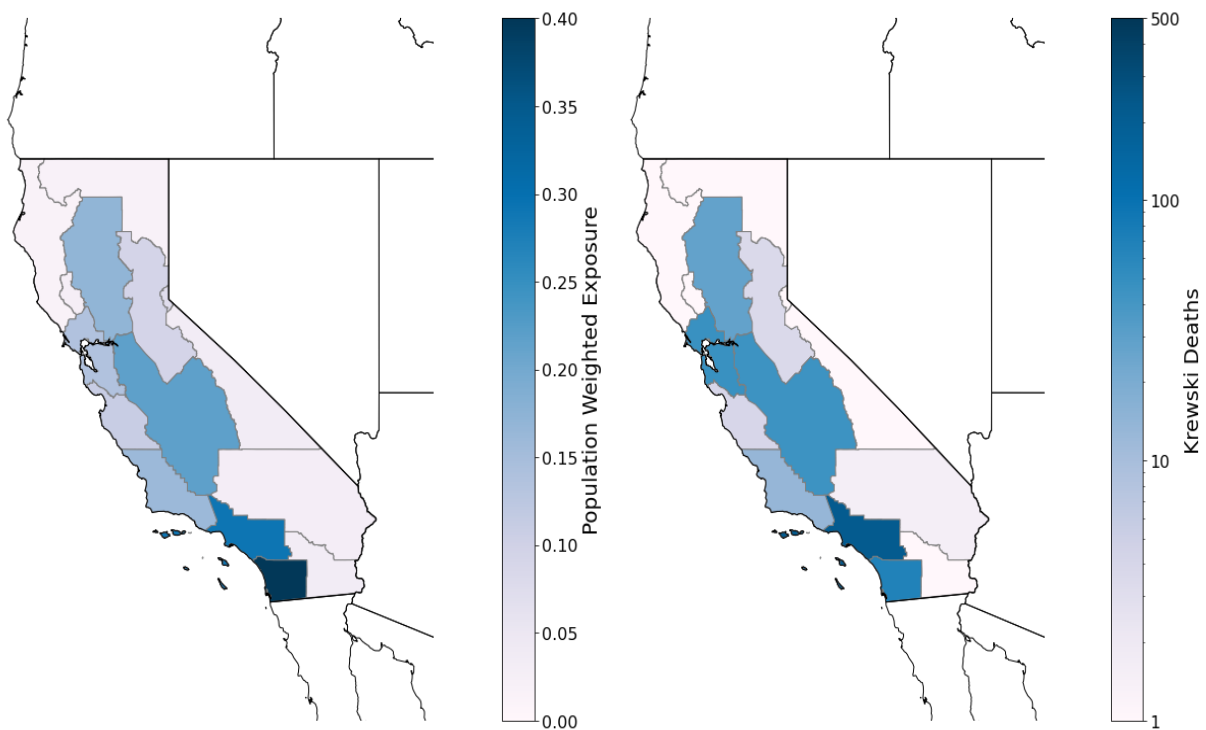


Figure 4.7: Population-weighted exposure (left) and reduced premature deaths associated with secondary $\text{PM}_{2.5}$ exposure (right) in each air basin due to decreases in secondary $\text{PM}_{2.5}$ concentration in 2040 in comparison to the 2021 baseline.

Table 4.2 summarizes the forecasts of public health benefits to the population of each air basin, showing avoided premature deaths associated with the reduction in secondary PM_{2.5} concentrations. This summary includes deaths from all causes, as well as associated premature mortality for more specific conditions: cardiopulmonary disease, ischemic heart disease, and lung cancer. The results reveal that the South Coast Air Basin will see the highest number of avoided premature deaths by a large margin, primarily due to the high population of the air basin. For perspective, one previous study found that the PM_{2.5}-associated mortality in California ranged from 12,700 to 26,700 deaths per year (with 47% from in-state anthropogenic emissions) in 2012, and another study estimated 8,300 premature deaths attributable to PM_{2.5} and O₃ formed from NO₂ vehicle emissions in California in 2016.^{147,148} While the total number of excess deaths in the San Joaquin Valley and San Francisco Bay Area are similar as seen in the previous figure, the San Joaquin Valley has a much lower overall population in comparison.

Table 4.2: Annual avoided premature deaths associated with the secondary PM_{2.5} concentration difference between 2040 to 2021 for all causes, cardiopulmonary disease, ischemic heart disease, and lung cancer.

Air Basin	All Cause	Cardiopulmonary Disease	Ischemic Heart Disease	Lung Cancer
South Coast	239	410	1046	461
San Francisco Bay	50	85	205	95
San Joaquin Valley	49	82	200	92
San Diego County	75	129	330	145
Sacramento Valley	30	51	123	57
South Central Coast	14	23	56	26
Mojave Desert	1.7	2.7	6.2	3.0
North Central Coast	4.2	7.0	16	7.7
Salton Sea	1.1	1.8	4.1	2.0
Mountain Counties	3.8	6.3	15	7.0
North Coast	0.4	0.7	1.6	0.8
Northeast Plateau	0.2	0.3	0.6	0.3
Great Basin Valleys	0.2	0.3	0.6	0.3
Lake County	0.2	0.3	0.7	0.3
Lake Tahoe	0.2	0.4	0.8	0.4

When examining the two most populated urban areas in California, a consistent pattern is evident: large decreases in NO_x concentrations in densely populated regions are accompanied by relatively small decreases in secondary PM_{2.5} in those same areas. Larger decreases in secondary PM_{2.5} relative to the population are seen in the San Joaquin Valley and San Diego County. The results indicate that while the vehicle-sourced NO_x concentrations closely mirror the spatial patterns of emissions, the impacts of these changes on secondary PM_{2.5} are anticipated to be larger at locations further downwind from the source. Potential causes are significant transport of NO_x away from cities occurring before particulate nitrate can form and a higher abundance of ammonia further from urban centers, allowing for increased rates of formation of secondary PM_{2.5}. The observed pattern may, in part, also reflect the limitations of the reduced complexity model employed. Future investigations using updated inputs in InMAP or employing a comprehensive chemical transport model would provide a deeper understanding of the underlying processes contributing to the observed patterns.

The absence of clear patterns in exposure changes by race and ethnicity emphasizes the necessity for more targeted strategies to address disparities in the health burden of air pollution. While the decreases in vehicle NO_x emissions are good for overall health in California, the projected decreases alone are insufficient to reduce the disparity in exposure between white and minority communities. Reductions in race-related health burden disparities from air pollution exposure could be achieved by targeting specific regions for accelerated rollout of vehicle electrification efforts, shifting the emission control programs to focus on regions with high PM_{2.5} formation, or reducing emissions of other PM_{2.5} precursors such as NH₃ from agriculture. The results underscore the need for spatial nuance when crafting future policy changes to maximize decreases in exposure to secondary PM_{2.5} which may be spatially distinct from the locations with the highest emissions of NO_x. For instance, large decreases in emissions expected to occur in the San Francisco Bay area may have a slight benefit to health outcomes due to secondary PM_{2.5} exposure reductions in the pollution-burdened San Joaquin Valley nearby where there is a high density of disadvantaged communities.

This research mainly addresses changes in air pollution exposures and excess mortality associated with secondary PM_{2.5}. However, additional reductions in other primary pollutants from vehicle exhaust are likely to result from vehicle NO_x reduction efforts and additional analysis using spatial disaggregation of disadvantaged and historically redlined communities is necessary to understand the full extent of health impacts from an environmental justice lens. These reductions will likely sit closer to the sources, similar to the patterns reported here for NO_x, and primarily reduce health impacts on near-roadway communities. Future studies incorporating primary PM_{2.5} directly emitted in exhaust (e.g., soot and oil particles) and produced through brake and tire wear will provide a more comprehensive view of the health benefits of vehicle emission control efforts. Reductions in vehicle emissions, especially NO_x, are also anticipated to affect other secondary pollutants such as ozone. The magnitude and spatial distribution of these additional air quality and health benefits should be studied further to inform targeted and effective policies for improving air quality and public health in California.

Chapter 5: Conclusions

5.1 Summary of Major Findings

Vehicle emissions of NO_x significantly contribute to urban air pollution through their role in the formation of secondary pollutants like ozone and PM_{2.5}, meaning that understanding their source contribution is critical to human and environmental health. However, quantifying vehicle emissions is challenging due to the range of engines of varying ages, emission control technology capabilities, and fuel types. There is the additional challenge of quantifying the spatial and temporal variability of mobile sources. Current understanding of the distribution and magnitude of the effects is limited and previous attempts at characterization of emissions via remote sensing observations have revealed large uncertainties in past and present emissions estimates. Without knowing the degree to which previous policy changes and technological advancements have affected air quality up to this point, future emission forecasts are difficult to rely on to guide management strategies and target specific source sectors. This body of research covers historical state and national NO_x emission trends, the creation and evaluation of a high-resolution vehicle emissions inventory, and an exploration of how future vehicle fleet compositions will affect spatial patterns of pollutants and human health effects.

Chapter 2 illuminated the evolving landscape of vehicle NO_x emissions, spanning the period from 1990 to 2020, both nationally and within the state of California. This analysis formed the backdrop for subsequent investigations into how these emissions manifest spatially in the present and how they will change moving forward. Separate emission factors and total emissions trend lines for the US and California were determined to account for the rapid turnover and replacement of heavy-duty diesel engines due to state mandates in California. The emission factor trend for gasoline engines indicates diminishing returns at present and small decreases in emissions year to year. This research extends previous fuel-based inventories by evaluating the light- and medium-duty diesel engines separately. The NO_x emissions from this sector are found to be decreasing more slowly than the heavy-duty diesel and gasoline vehicle categories. The light/medium-duty diesel vehicle category contribution to NO_x emissions in urban areas has been overlooked to some degree in current regulatory inventories.

The overall reductions in on-road vehicle NO_x emissions nationwide and in California are approximately 69 and 71%, respectively, between 1990 and 2020. Because of the high rate of replacement of older heavy-duty diesel engines in California, reductions in NO_x emissions have accelerated over the last decade. Diesel NO_x emissions in California are estimated to have decreased by 48% since 2010 in comparison to 32% nationally over the same time period. Emission inventory results from this research were also compared to the national EPA MOVES and California EMFAC models. MOVES predicts higher NO_x emissions overall for all years for gasoline engines and a steeper decrease in emissions compared to the fuel-based inventory. The EMFAC model is largely in agreement with the fuel-based inventory for the gasoline vehicle category. EMFAC estimates are higher for diesel engines before the mid-2010s but have a similar trend to the fuel-based inventory from 2010-2020.

Chapter 3 of this dissertation focuses on the development and evaluation of a high-

resolution fuel-based emission inventory for Los Angeles for the period of June 2021, aiming to gain a better understanding of source contributions in the domain. Unlike existing inventories used in other air quality modeling studies, the inventory developed in this research provides unusually high-resolution grid cells of $1.3 \times 1.3 \text{ km}^2$ and incorporates the new light- and medium-duty diesel engine category. The inventory was used as input to a nested grid configuration of the WRF-Chem model to predict NO_2 concentrations. Model results were compared to the TROPOMI satellite, vertical profiles from the RECAP-CA aircraft campaign, and ambient sensors. The focus of the comparison with satellite data was on weekday-weekend differences in emissions and resulting concentrations. Model performance on both weekdays and weekends was consistent and indicated appropriate day-of-week scaling of the emissions.

A key finding is that on-road vehicles are still the dominant contributor to NO_x emissions in the Los Angeles area, and were responsible for 55-72% of emissions, compared to an estimate of 43% of total NO_x emissions from the South Coast Air Quality Management District. Most of this difference stems from uncertainties in emissions from the gasoline and light-/medium-duty diesel categories. The fuel-based inventory has NO_x emissions that are 1.5 ± 0.4 , 2.8 ± 0.6 , and 1.3 ± 0.7 times the corresponding EMFAC model estimates for gasoline, light-/medium-duty diesel, and heavy-duty diesel respectively. These differences could be due to several factors including slow absolute progress in reducing emission factors from gasoline engines and slow turnover of high-emitting light- and medium-duty diesel engines. Leveraging WRF-Chem and satellite data, Chapter 3 of this dissertation provided a granular understanding of the spatial distribution of emissions at a local scale. The findings not only shed light on the current state of vehicular emissions in the Los Angeles area, but also have laid new groundwork for developing and verifying emission inventories in other urban areas, which in turn could support more accurate air quality modeling and more effective and better-targeted air pollution control efforts.

The culmination of the research and the health and environmental justice implications of this work are described in Chapter 4, where the high-resolution motor vehicle emission inventory was expanded from the urban Los Angeles area to the statewide scale. The new high-resolution inventory was used to project spatial distributions of the expected changes in secondary $\text{PM}_{2.5}$ and associated health effects across all of California. These projections offer insights into potential regional challenges and emphasize the urgency of proactive measures to safeguard public health. The high-resolution NO_x emission inventory was expanded first to a statewide spatial extent and annual average conditions for a baseline year (2021), and then to forecast future patterns of NO_x emissions for future years (2030 and 2040). On-road vehicle NO_x emissions are projected to decrease relative to baseline conditions by 38% in 2030 and by 55% in 2040. In comparison to EMFAC model predictions, the fuel-based inventory has 63% higher emissions overall in 2021. This gap between emission estimates shrinks in future projections but persists largely due to differences in emission estimates for light- and medium-duty diesel vehicles.

Using the fuel-based emission inventory as input in InMAP, several key patterns emerge. While the largest NO_x concentration decreases are close to sources such as major highways and dense urban centers, the largest secondary $\text{PM}_{2.5}$ concentration decreases are distinctly separated in space from emissions. For the two most populous areas of the state, the most significant decreases in secondary $\text{PM}_{2.5}$ are seen further inland for the San Francisco Bay Area towards the

Central Valley and further south towards San Diego instead of within the LA basin. This suggests that regional-scale transport of $\text{PM}_{2.5}$ and its precursors is significant and that there may be faster NO_x to particulate nitrate conversion rates further from urban centers. This is important to consider for air quality management strategies, as the health benefits may not be co-located with where the largest decreases in emissions are taking place. Results also indicate no significant pattern in populated-weighted exposure by race with regard to vehicle emissions decreases, suggesting that while exposure is decreasing overall, there is a need to have better-targeted strategies. Whether these strategies target specific regions or emission source sectors, the current trajectory is not likely to reduce the existing racial disparities and inequity in exposure to air pollution. While this study focuses on reductions of NO_x and secondary particulate nitrate, the decrease in vehicle emissions will likely have the added benefits of decreasing co-pollutants and other secondary products such as ozone.

5.2 Recommendations for Future Research

The results of this research have led to the identification of additional questions that should be addressed to better understand the vehicle contribution to NO_x concentrations and how emission control technologies affect air quality and health in the real world. Such studies would improve the understanding of the overall NO_x budget. The evolution of vehicle types and categories will need to be tracked in an ongoing manner using updated methods. The on-road vehicle fleet is undergoing rapid progress through the implementation of emissions control technologies and vehicle electrification. There is potential for increased emissions as well, such as through emissions control system degradation and the prospect of growth in fuel sales outpacing emission factor reductions. By offering a comprehensive understanding of historical trends, present conditions, and projections of future NO_x emissions, the results laid out in the preceding chapters improve the understanding of the largest and most critical sector for improving air quality in cities. This research provides a framework for high-resolution vehicle emissions mapping and air quality modeling that can be used in further applications to understand air pollution problems elsewhere in the US and worldwide.

5.2.1 Evaluating spatial and temporal trends in emissions using new satellites

While general conclusions can be made for how effective reducing emissions is on an annual average timescale, NO_x concentrations and secondary pollutants formed from NO_x have diurnal, weekly, and seasonal variations. Additional studies should highlight the importance of short-term exposure to very high levels of air pollution. This effect is not fully captured in the existing research, especially for $\text{PM}_{2.5}$ where only annual average concentrations were modeled. The newly launched TEMPO satellite with a native resolution of around $2 \times 3 \text{ km}^2$ provides a path forward to uncovering more information about source sectors of NO_x . The high-resolution gridding of the emission inventory and model runs presented in this dissertation anticipates this transition to higher-resolution satellite data. Similar comparisons could be used to constrain NO_x emissions more effectively over larger areas such as the continental US. The TEMPO satellite is geostationary with a field of view that remains fixed over North America. TEMPO is therefore able to provide data at multiple daytime hours whereas TROPOMI satellite data used in the present work only provides a single fly-over at around 1:30 PM daily. While the weekday-

weekend pattern of the on-road sector is a key point of interest in this research, taking a closer look at how the diurnal pattern of modeled NO₂ concentrations compares to satellite data could also reveal key information about how accurately NO_x emissions have been estimated.

5.2.2 Closer examination of the NO_x emissions in the light and medium-duty diesel vehicle and off-road mobile source sectors

One of the major findings of this research is the underestimation of NO_x emissions from light- and medium-duty diesel vehicles. This sector includes a wide range of vehicles of varying sizes and uses and is poorly understood. Available emission measurements may not be representative of the entire fleet. As gasoline engines have achieved very low NO_x emission factors and heavy-duty diesel trucks have fully adopted advanced SCR-based emission control systems, the emissions from light- and medium-duty diesel engines are becoming increasingly important. Developing better methods for modeling the boom in e-commerce and warehousing is recommended. While much of the information about warehouse locations and related drayage truck and delivery vehicle trips is either outdated or difficult to access publicly, some of the model-measurement mismatch seen in Southern California could be due to the rapid expansion of warehouses and associated truck traffic traveling to and from inland areas. Another source of uncertainty that arose in this research was emissions from off-road and marine engines. Better quantification of both the spatial patterns and magnitude of NO_x emissions from these sources is needed.

5.2.3 Expanding the fuel-based emissions analysis to other major cities

Los Angeles is a key area of interest in relation to air pollution and on-road vehicle emissions due to a high reliance on car transportation, a high volume of freight shipments originating from the Ports of Los Angeles and Long Beach, and the lack of significant emissions from other traditional sources such as fossil-fueled power plants and heavy industry. Los Angeles has notable differences from other major US cities, including unique meteorology due to its coastal location and mountain ranges enclosing the air basin. A similar analysis of emissions and air quality is recommended for the San Francisco Bay Area and nearby cities in the Central Valley (e.g., Sacramento and Fresno) to investigate how emissions vary regionally. These cities follow the same state mandates for the advancement of mobile source emission controls and the adoption of new vehicle technologies such as electric powertrains and fuel cells. Similar analyses outside of California would help determine source contributions to NO_x emissions in other major US metropolitan areas with more non-road and industrial emission sources.

5.2.4 Evaluating future emission scenarios using chemical transport models and updated versions of InMAP

While Chapter 4 gives an overview of the magnitude and spatial distribution of health impacts associated with expected NO_x emission reductions in future years, limitations of the current InMAP modeling approach leave room for further research. Despite significant computational costs, running future emission scenarios using a fully coupled weather and

chemistry model for an entire year case would be useful in recalibrating and extending the results of InMAP. Using a more recent WRF-Chem run as the basis for predictive studies using InMAP is recommended due to expected changes in NO_x lifetimes and atmospheric chemistry in urban areas as NO_x emissions continue to decrease. In addition to updates to the current meteorological parameterizations, more recent census data from 2020 should be incorporated when available to study further racial and socioeconomic differences in exposure to air pollution that are expected as vehicle emissions continue to change over time.

5.2.5 Consideration of other key secondary pollutants, especially ozone.

This modeling and evaluation project occurred during simultaneous efforts by collaborators at NOAA to improve the parameterization of VOCs and photolysis schemes in the WRF-Chem setup. While this research focuses on NO_x only, a more holistic evaluation of the emissions inventory including ozone and formaldehyde as a proxy for NMVOCs would give critical insight into the current chemical regime of Los Angeles. This analysis could potentially reveal more about how the on-road sector contributes to ozone exceedance events. Modeled ozone concentrations could potentially be used to look at exposure and health effects. Additionally, InMAP runs could integrate the primary PM_{2.5} emissions from exhaust, brake, and tire wear that are beyond the scope of the current NO_x-focused studies. An examination of the health impacts of ozone and primary PM_{2.5} attributable to vehicle emissions would complement the analysis done in Chapter 4 and provide a more complete view of all the health benefits of the transition to a low-emissions fleet.

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